Study of particulate matter removal mechanism by using non-thermal plasma technology

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Abstract

Numbers of diesel engines in both stationary and mobile applications are increasing nowadays. Diesel engines emit lower Hydrocarbon (HC) and Carbon monoxide (CO) than gasoline engines. However, they can produce more nitrogen oxides (NO_x) and have higher particulate matter (PM). On the other hand, emissions standards are getting stringent day by day due to considerable concerns about unregulated pollutants and particularly ultrafine particles deleterious effect on human health. Non-thermal plasma (NTP) treatment of exhaust gas is known as a promising technology for both NO_x and PM reduction by introducing plasma inside the exhaust gas. Vehicle exhaust gases undergo chemical changes when exposed to plasma. In this study, the PM removal mechanism using NTP by applying high voltage pulses of up to 20 kV_{pp} with a repetition rate of 10 kHz are investigated. It is found that, voltage increase not necessarily has a positive effect on PM removal in diesel engine emissions.

Keywords: Non-thermal plasma, Particulate matter removal, Dielectric barrier discharge, Pulsed power supply

1. Introduction

Diesel engines have been employed in variety of industries and their applications are growing rapidly. Despite the large number of diesel engine applications, little has been done to reduce the large amounts of pollutants caused by diesel engines. According to the negative effects of NO_x and PM on health, governmental legislations for permissible exhaust emission standards are becoming more and more stringent day by day.

Up to now, several technologies have been applied for NO_x and particulate treatment of diesel engines. For example, the selective catalyst reduction (SCR) method is one of the common technologies for NO_x reduction of automobile and stationary engines [1]. There are some problems in using SCR catalysts such as the possibility of ammonia leakage, catalyst poisoning, catalyst discharge under the high temperature condition or under influence of Sulphur and need for construction of urea solution stations. Diesel particulate filters (DPF) have been used widely for particulate matter removal in diesel engines [2]. However, there are some drawbacks in using DPF such as pressure drop inside the exhaust gasses due to the PM deposition. This deposition can cause filter choking so filter regeneration at high temperature about 600 °C is required. These effects cause more fuel consumption which is not appropriate for emission production and fuel economy.

Electrical discharge plasma technique appears to be a very promising method for both NO_x and PM reduction [3]. It is composed of free electrons, ions, radicals, atoms, and molecules in various states of excitation. In Non-thermal plasma, the majority of electrical energy goes into the production of high-energy electrons rather than into gas heating [4]. The discharge achieves non-thermal conditions through the production of short-lived micro discharges. The short lifetime of the micro-discharges is achieved by applying very-short high-voltage pulses [5]. The intent of using non-thermal plasma is to selectively transfer the input electrical energy to the electrons which would generate free radicals through collisions and promote the desired chemical changes in the exhaust gas [6]. In other words, when plasma is introduced in the exhaust gases, oxidation processes will be started. NO_x, unburned hydrocarbons, carbon monoxide and particulate matter (PM) will be oxidized. In spite of NO_x reduction to N_2 and O_2, plasma treatment of exhaust gases is more related to NO oxidation to NO_2 [7]. The plasma is believed to show potential to improve catalyst selectivity and removal efficiency. Moreover plasma can oxidize PM in diesel exhaust gases.

The majority of studies in this area are related to the effect of plasma on NO_x removal [8-10]. However, PM removal efficiency of plasma has not been fully investigated. In 2000,Thomas et.al [3], considered NTP for emission treatment of exhaust gases. They used a packed bed reactor and found that by increasing the residence time selectively, when using a packing material into the NTP reactor, the level of oxidation of species may be decoupled from the energy deposition into the exhaust gas. Their packed plasma system removed 99.9% of PM of average diameters around 60 nm. Yeo et.al [11] used a plasma reactor with catalyst to treat exhaust gas from a gasoline engine to reduce PM emissions. In their experiments, they considered PM diameter ranges of 0.3 to 5 μm in a discrete manner. Their result indicated that PM removal efficiency ranged approximately from 25 to 75%. Suzuki et.al [12]used carbon black particles (CBP) to simulate the diesel engine particles in their experiments. They could remove about 80% of the CBP by introducing plasma. Song et.al considered the PM, hydrocarbons (HC) and NO_x abatements characteristics in 2009 [13]. They studied the effect of peak voltage, frequency and engine load on contaminant removal by using a dielectric barrier discharge (DBD) reactor consisting of two concentric quartz tubes. They showed that by increasing voltage at each frequency level, discharge power increases. But there is an optimum for frequency about 15.5 kHz regarding the discharge power. They found PM, HC and NO_x abatements obviously increase with increasing the applied voltage. Approximately for all voltage levels, the maximum of discharge power and consequently maximum of removal efficiency obtained on about 15 KHz. In 2010 Okabo et.al[14] used packed pellets of $BaTiO_3$ for simulation of non-thermal plasma for PM and NO_x removal. They confirmed that carbon soot is removed under oxygen poor conditions and simultaneous removal of PM and NO_x happened.

The aim of this paper is to study the effect of NTP on the PM removal mechanism from the exhaust gases of a real diesel engine. In this regard, the hypothesis of gas to particle change by using NTP technology was evaluated. Regarding the PM after treatment systems, PM mass reduction and PM size distribution was considered simultaneously. The experiments are conducted using a DBD reactor which has been designed and fabricated for this purpose. Employing a pulsed power supply developed based on the power electronics technology [15-17], provided the ability to sustained NTP with in the DBD reactor and possibility of applying different voltage levels [18].

2. Experimental setup

A dielectric barrier discharge reactor is designed for the experiments. It consists of two concentric quartz tubes. Both tubes are 400 mm long and have the wall thickness of 1.5 mm. The outside diameter of inner and outer quartz tubes are 20mm and 25mm respectively. Exhaust gas passes through the gap between these two quartz tubes. Based on pre-designed geometry the discharge gap will be 1 mm. The internal electrode is a copper cylinder and the external electrode is made by a copper mesh that wraps the exterior part of the DBD. The discharge length for primary experiments was 100 mm. Fig .1 shows the designed reactor and employed pulsed power supply [18]. By applying high voltage pulses (see Fig. 2), plasma discharge will happen between these two electrodes. The dielectric has two functions of limiting the charge transferred by an individual

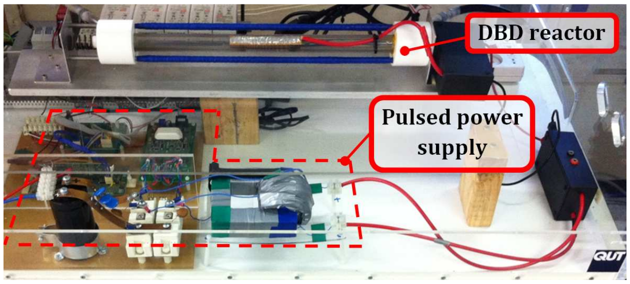

Fig. 1. Plasma reactor and electrical setup.

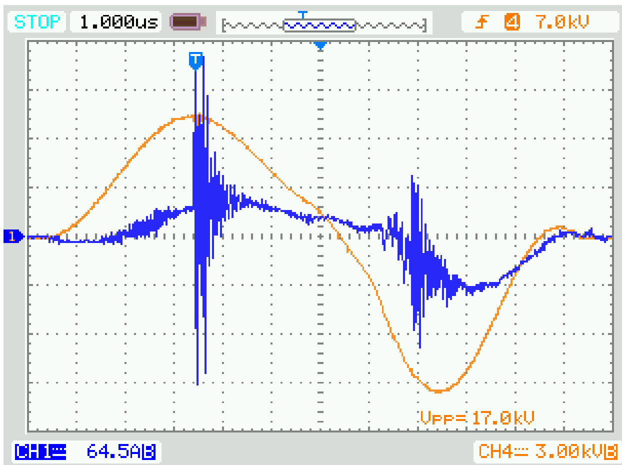

Fig. 2. Typical measured waveforms of pulsed voltage and current of the DBD reactor.

micro-discharge and spreading the micro-discharge over the electrode surface tube [19]. Both tubes are fixed by two Teflon caps at each ends and exhaust enter the reactor by the angle of 45 degree and flow throughout the gap and leave the reactor with same angle.

A schematic diagram of experimental setup is shown in Fig. 3. Experiments were conducted on a modern turbo-charged 6-cylinder Cummins diesel engine (ISBe220 31). A pulsed power supply is used to induce high voltage between the two dielectric layers. The applied voltages are in 15-20 kV_{pp} range at frequency of 10 kHz. When the breakdown voltage is reached a discharge will be started and ionization occurs and an electrical current starts flowing into the gas.

Particle number distributions are measured with a Scanning Mobility Particle Sizer (SMPS) consisting of a TSI 3080 classifier, which pre-

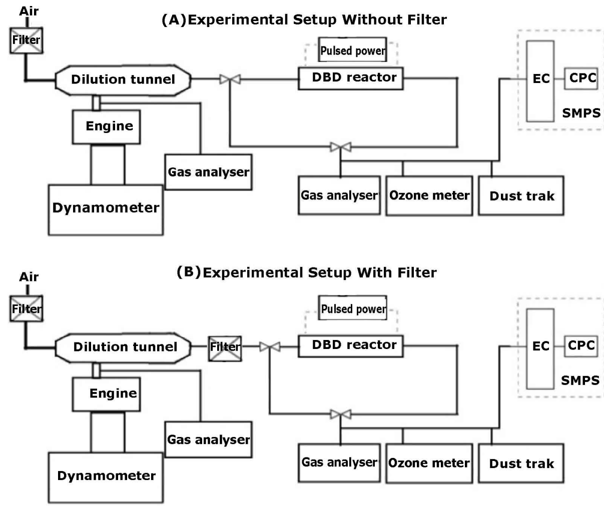

Fig. 3. Schematic diagram of experimental setup: (A) without filter-(B) with filter.

selects particles within a narrow mobility. Gaseous emissions are measured with CAI 600 series gas analyzer and particulate mass emissions are measured with a TSI 8530 Dust-Track II.

3. Results and discussion

3.1. Plasma effect on particle size distribution

For studying the effect of plasma on emission treatment of exhaust gases, some initial experiments have been carried out considering the emissions of Cummins diesel engine. In all experiments engine speed and load kept constant at 40 kW (25% load) and 2000 rpm, respectively. A portion of raw exhaust gas directly from an iso-kinetic sampling port of the tailpipe has been diluted with air and passed through the reactor. The dilution ratio was constant at about 10 in all experiments. The plasma removal effect was considered at three different voltage levels at 15, 17 and 20 kV_{pp}, which are correspond to different discharge powers. First, the maximum voltage level has been applied to make sure that the breakdown voltage has been met and plasma happened. Emission concentrations were measured in reactor inlet (reactor inlet no pulse) and reactor outlet (reactor outlet no pulse) without applying any pulse voltage to see the rate of deposition on the reactor surface. Then, pulse voltage was

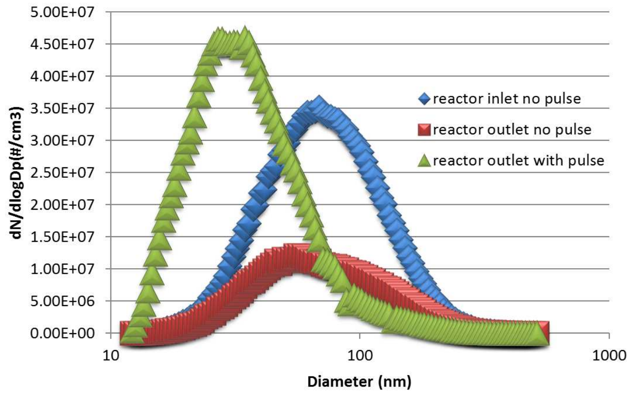

Fig. 4. Particle size distribution (2000rpm, 25% Load, $20kV_{pp}$ and 10 kHz)

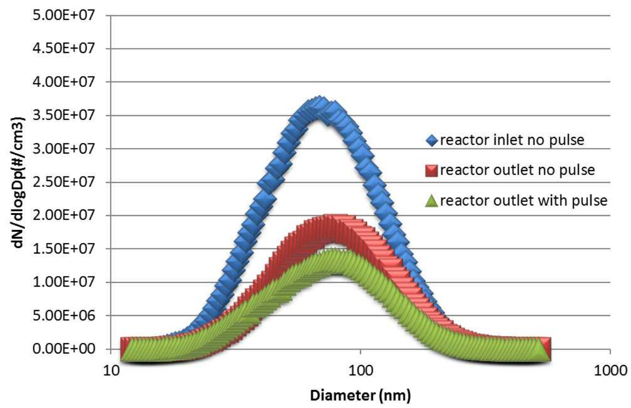

Fig. 5. Particle size distribution (2000rpm, 25% Load, $17kV_{pp}$ and 10 kHz)

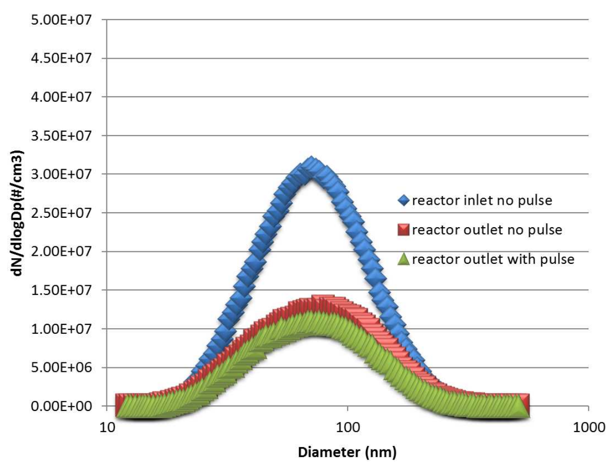

Fig. 6. Particle size distribution (2000rpm, 25% Load, $15kV_{pp}$ and 10 kHz)

applied to produce plasma and particle size distribution was measured in reactor outlet (reactor outlet with pulse). This procedure was repeated for the rest of measurements. The effect of plasma on particle size distribution has been shown in Figs. 4, 5, and 6.

PM diameters, whose removal is being targeted, are in the range of 10nm to 500nm. As it is shown, particle size distributions are different under varied voltage levels. Fig. 6 shows that particle size distribution changes slightly by applying 15 kV_{pp}. As depicted in Fig 5 the PM removal is improved by increasing the applied voltage up to 17 kV_{pp}. When the voltage level goes up to 20 kV_{pp}, larger particles have been removed more; however, many smaller particles have been produced. Therefore, regarding the particle size distribution, increasing the voltage level above 17 kV_{pp} has adversely affect the system performance.

As it can be seen at 20 $kV_{pp,}$ the number of tiny particles was increased dramatically. Two possibilities can be considered for this phenomenon:

- Plasma changes some part of exhaust gas to particles.
- Larger particles break to smaller particles or probably some condensation happens.

For studying the first theory, a PM filter was added in reactor inlet and exhaust gases passed through this filter before entering the reactor. The schematic diagram of this setup has been illustrated in Fig. 3B and the results have been summarized on three following figures.

As shown in Figs. 7, 8 and 9, by adding this filter, almost all particles have been removed from the exhausts gas before entering the DBD reactor. Just a small portion of particles have not been trapped by the filter. After introducing plasma, most of the un-trapped particles are removed when the voltage levels are 15 kV_{pp} and 17 kV_{pp}. However, as Fig. 9 depicted, the effect of plasma at 20 kV_{pp} on particle size distribution is different. Without applying any pulse, the number of particles is very small. But, by applying 20 kV_{pp} and introducing plasma inside the reactor number of small particles was massively increased. This result indicates the occurrence of gas to particle reactions at this voltage level.

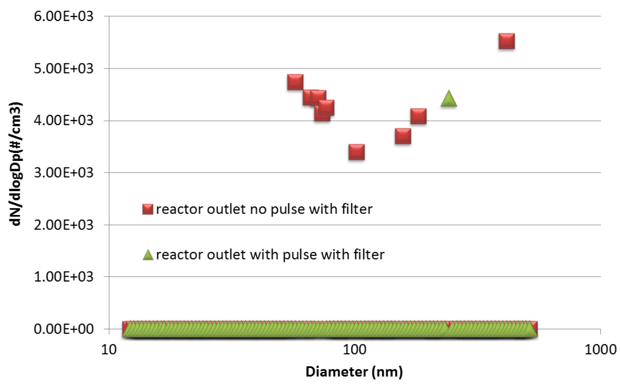

Fig. 7. Particle Size Distribution with filtering (2000rpm, 25% Load, 15kV$_{pp}$ and 10 KHz).

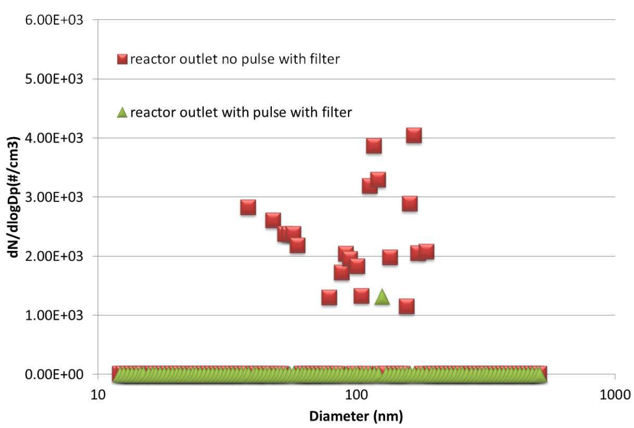

Fig. 8. Particle Size Distribution with filtering (2000rpm, 25% Load, 17kV$_{pp}$ and 10 KHz).

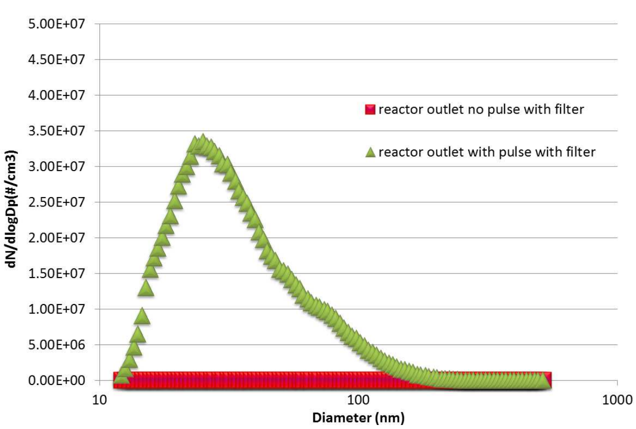

Fig. 9. Particle Size Distribution with filtering (2000rpm, 25%Load, 20kV$_{pp}$ and 10 KHz).

Table 1. The concentration of PM before and after the treatment at different voltages

Operating condition / Filtering	20 kVpp @10 kHz		17 kVpp @10 kHz		15 kVpp @10 kHz	
	No	Yes	No	Yes	No	Yes
Reactor Inlet PM Concentration ($\frac{mg}{m^3}$)	**4.56**	**-**	**4.26**	**-**	**5.14**	**-**
Reactor Outlet No Pulse PM Concentration ($\frac{mg}{m^3}$)	**2.84**	**0.012**	**3.68**	**0.01**	**4.37**	**0.013**
Reactor Outlet By-Pulse PM Concentration ($\frac{mg}{m^3}$)	**2.56**	**0.084**	**2.62**	**0.01**	**3.74**	**0.013**
Plasma PM Removal Efficiency (%)	**43.9**	**-**	**38.6**	**-**	**27.1**	**-**

Therefore, some part of increase on PM numbers obtained on Fig. 4 could be due to the gas to particle changes.

3.2 Particle mass effect by introducing plasma

In this section, the effect of plasma on particle mass reduction is presented. Three different voltage levels have been considered same as previous section. All results have been summarized in Table 1. The PM mass reduction without using any filter for 20 kV$_{pp}$, 17 kV$_{pp}$ and 15 kV$_{pp}$ are 43.9 %, 38.6% and 27.1%, respectively. The maximum PM mass reduction has been obtained when the voltage level is 20 kV$_{pp}$. Also, a reasonable mass reduction can be obtained around 40 % by applying 17 kV$_{pp}$ without any increase in ultra-fine particle numbers.

When a filter is added at reactor inlet, the PM concentration is very low due to the filter trapping compare to the PM concentration without using any filter. Particle mass before and after applying a pulse were measured to clarify the amount of gas to particle changes. Without applying high voltage pulses, the particle mass concentration in the reactor outlet is $0.012\frac{mg}{m^3}$, $0.01\frac{mg}{m^3}$ and $0.013\frac{mg}{m^3}$ for three different measurements. However, by applying

20 kV_{pp} PM mass increases to $0.084\frac{mg}{m^3}$. This shows that some part of the gaseous pollutants change to particles due to the plasma effect. At 17 kV_{pp} and 15 kV_{pp}, there is no increase in particle mass by introducing plasma. Further studies are required to completely understand this effect.

4. Conclusion

In this paper, the effect of non-thermal plasma on PM removal mechanism and PM size distribution at different voltage levels has been studied. NTP was generated and controlled at different operating point by using a pulsed power supply. A DBD reactor with two dielectrics was employed in order to generate plasma. The results showed the ability of proposed system in decreasing PM concentrations. Furthermore, it is found that voltage increase not necessarily has a positive effect on PM removal in diesel engine emissions. This has been validated based on the hypothesis of gas to particle change by using NTP technology.

References

[1] C. K. Narula, C. S. Daw, J. W. Hoard, and T. Hammer, "Materials Issues Related to Catalysts for Treatment of Diesel Exhaust," *International Journal of Applied Ceramic Technology*, vol. 2, pp. 452-466, 2005.

[2] T. V. Johnson, "Vehicular Emissions in Review," *V121-3EJ*, vol. 5, pp. 216-234, 2012.

[3] S. E. Thomas, A. R. Martin, D. Raybone, J. T. Shawcross, K. L. Ng, P. Beech, *et al.*, "Non Thermal Plasma Aftertreatment of Particulates – Theoretical Limits and Impact on Reactor Design," presented at the International Spring Fuels & Lubricants Meeting & Exposition, Paris, France, 2000.

[4] B. M. Penetrante, *Non-Thermal Plasma Techniques for Pollution Control: Part A: Overview, Fundamentals and Supporting Technologies. Part B: Electron Beam and Electrical Discharge Processing*: Springer, 1994.

[5] K. Yan, H. Hui, M. Cui, J. Miao, X. Wu, C. Bao, *et al.*, "Corona induced non-thermal plasmas: Fundamental study and industrial applications," *Journal of Electrostatics*, vol. 44, pp. 17-39, 1998.

[6] "Plasma Exhaust Treatment," ed, 2000, p.http://www.dieselnet.com/tech/plasma.html.

[7] B. S. Rajanikanth, S. Das, and A. D. Srinivasan, "Unfiltered Diesel Engine Exhaust Treatment by Discharge Plasma: Effect of Soot Oxidation," *Plasma Science & Technology*, vol. 6, pp. 2475-2480, 2004.

[8] S. Mohapatro and B. S. Rajanikanth, "Cascaded cross flow DBD-adsorbent technique for NOX abatement in diesel engine exhaust," *Dielectrics and Electrical Insulation, IEEE Transactions on*, vol. 17, pp. 1543-1550, 2010.

[9] T. Vinh, S. Watanabe, T. Furuhata, and M. Arai, "Fundamental study of NOx removal from diesel exhaust gas by dielectric barrier discharge reactor," *Journal of Mechanical Science and Technology*, vol. 26, pp. 1921-1928, 2012/06/01 2012.

[10] J. Jolibois, K. Takashima, and A. Mizuno, "Application of a non-thermal surface plasma discharge in wet condition for gas exhaust treatment: NOx removal," *Journal of Electrostatics*, 2012.

[11] D. Ye, D. Gao, G. Yu, X. Shen, and F. Gu, "An investigation of the treatment of particulate matter from gasoline engine exhaust using non-thermal plasma," *Journal of Hazardous Materials*, vol. 127, pp. 149-155, 2005.

[12] K. SUZUKI, N. TAKEUCHI, K. MADOKORO, C. FUSHIMI, S. YAO, Y. FUJIOKA, *et al.*, "Removal Properties of Diesel Exhaust Particles by a Dielectric Barrier Discharge Reactor," *Analytical Sciences*, vol. 24, pp. 253-256, 2008.

[13] C.-L. Song, F. Bin, Z.-M. Tao, F.-C. Li, and Q.-F. Huang, "Simultaneous removals of NOx, HC and PM from diesel exhaust emissions by dielectric barrier discharges," *Journal of Hazardous Materials*, vol. 166, pp. 523-530, 2009.

[14] M. Okubo, T. Kuroki, K. Yoshida, and T. Yamamoto, "Single-Stage Simultaneous Reduction of Diesel Particulate and NOx Using Oxygen-Lean Nonthermal Plasma Application," *Industry Applications, IEEE Transactions on*, vol. 46, pp. 2143-2150, 2010.

[15] P. Davari, F. Zare, and A. Ghosh, "A flexible solid-state pulsed power topology," in *Power Electronics and Motion Control Conference (EPE/PEMC), 2012 15th International*, 2012, pp. DS2b.12-1-DS2b.12-6.

[16] P. Davari, F. Zare, and A. Ghosh, "Parallel and series configurations of flyback converter for pulsed power applications," in *Industrial Electronics and Applications (ICIEA), 2012 7th IEEE Conference on*, 2012, pp. 1517-1522.

[17] P. Davari, F. Zare, A. Ghosh, and H. Akiyama, "High-Voltage Modular Power Supply Using Parallel and Series Configurations of Flyback Converter for Pulsed Power Applications," *Plasma Science, IEEE Transactions on*, vol. 40, pp. 2578-2587, 2012.

[18] M. Babaie, P. Davari, F. Zare, M. M. Rahman, H. Rahimzadeh, Z. Ristovski, *et al.*, "Effect of Pulsed Power on Particle Matter in Diesel Engine Exhaust Using a DBD Plasma Reactor," *IEEE Transactions on Plasma Science*, 2013 (Submitted).

[19] B. Eliasson, M. Hirth, and U. Kogelschatz, "Ozone synthesis from oxygen in dielectric barrier discharges," *Journal of Physics D: Applied Physics*, vol. 20, p. 1421, 1987.