Reducing Emissions of Polycyclic Aromatic Hydrocarbons and Greenhouse Gases from Engines Using a Novel Plasma-Enhanced Combustion System

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ABSTRACT

Plasma activation is an innovative concept for converting liquid or gaseous fuels into reactive species. However, there are few studies that focus on activating air to enhance combustion efficiency and reduce emissions of toxic pollutants such as PAHs. In this study, a plasma-enhanced combustion system was used to increase the amount of free radicals in intake air and enhance the combustion efficiency. The experimental results show that the reduction in pollutant emissions was enhanced with the increase in voltage, and that this also reduced the amount of fuel used. The optimum voltage of the plasma-enhanced system was 3200 V, and this reduced total PAHs by 20.7%, total BaPeq by 20.5%, CH4 by 50.0%, NMHC by 57.7%, THC by 57.1%, CO by 60.2%, NOx by 70.7%, CO2 by 19.8%, and PM by 42.8%, and achieved fuel savings of 12.1%.

Keywords: PAH; Emission; Plasma; Gasoline engine; Saving fuel; Greenhouse gases.

INTRODUCTION

The rapid depletion of global energy resources and increased concern on the impact of environmental issues due to fossil fuels make the requirements in thermal efficiency and low emissions for current engines more and more stringent. However, vehicles remain a major source of nitrogen oxides (NOx), carbon monoxide (CO), hydrocarbons (HC), particulate matter (PM) and polycyclic aromatic hydrocarbon (PAH) emissions, and many researchers have suggested that mobile sources are the major contributor in urban and suburban environments (Lin et al., 2006a, b, c; Tsai et al., 2010a, b; Wu et al., 2010; Hsieh et al., 2011a, b; Karavalakis et al., 2011; Kim et al., 2011; Li et al., 2011; Lin et al., 2011; Shen et al., 2011; Srivastava et al., 2011; Tsai et al., 2011; Yao et al., 2011; Cheng et al., 2012a, b; Lai et al., 2012; Liang et al., 2012; Liu et al., 2012; Pilusa et al., 2012; Tsai et al., 2012; Wang et al., 2012).

Some of PAHs are potential mutagens and carcinogens and are probably the major culprits in causing high lung cancer rate. In the past 30 years, many studies have suggested that PAHs are environmental immunosuppressive contaminants. PAHs, especially benzo(a)pyrene, not only injure the respiratory and immune system but also cause cell mutation and cancers such as lung and skin (Grevenynghe et al., 2003; Ji et al., 2009; Verhelst et al., 2009). Recently, much progress has been made in the control of combustion processes to limit the formation of environmentally harmful species such as hydrogen addition (Ji et al., 2010a, b; Wang et al., 2010; Zhao et al., 2010; Wang et al., 2011), biodiesel addition (Bueno et al., 2011; Lin et al., 2011; Valentino et al., 2011), methanol/ethanol addition (Berta et al., 2006; Graham et al., 2008; Abrantes et al., 2009; Najafi et al., 2009; Westphal et al., 2010; Lee et al., 2011; Ozsezen et al., 2011), ether oxygenate additives (Alex et al., 2007), premixed n-heptane flames (Li et al., 2007), and engine modification (Alkidas, 2007; Wel et al., 2007; Canakci, 2008).

Alkidas (2007) presented a critical review of the recent combustion advancements made in gasoline engines for the reduction of fuel consumption and engine emissions. For optimum combustion throughout their speed/load range, engines should be operated in three combustion modes: stratified-charge spark-ignited, homogeneous-charge spark-ignited, and homogeneous-charge compression-ignited. Key technologies to achieve this, with maximum reduction in fuel consumption and emissions, are spray-guided direct-injection system, flexible variable-valve actuation, and cylinder-pressure based engine controls (Alkidas, 2007). Wel et al. (2007) carried out on an electronically controlled fuel injection stoichiometric gasoline engine by using cold exhaust gas recirculation (EGR) and increasing compression ratio to improve fuel economy and reduce emissions. After the compression ratio of the engine was raised from 8 to
11.8, and EGR rate and air swirl ratio are optimized, the fuel economy is improved by 6.02%, and the NOx and (NOx + HC) emissions were decreased by 53.1% and 44.9%, respectively at full-load speed characteristics (Wel et al., 2007). Canakci (2008) concluded that the combustion efficiency increased with engine speed for the three boost pressure (101, 117, 138 kPa). For almost all engine speeds at the highest boost pressure, when start of injection timing was advanced which leads to less fuel stratification and lower combustion efficiency, the lowest NOx emissions were obtained by giving higher total unburned hydrocarbon emissions penalty (Canakci, 2008).

Nonthermal plasma activation is an innovative concept for converting liquid or gaseous fuels into reactive species. It involves prerecoking the fuel into the requisite reactive species by the application of atmospheric-pressure plasmas. In physics and chemistry, plasma is typically an ionized gas, and is usually considered to be a distinct state of matter in contrast to gases because of its unique properties. In plasma state, the electrical conductivity is very high. Also, electrons, ions, and neutrals can be distinguished by the sign of their charge so that they behave independently and can interact simultaneously with many others in many circumstances. All combustion processes rely on the breakdown of the fuel being burned into free radicals and other reactive species (Warnatz et al., 1996). It is this breakdown into reactive species that initiates the combustion process in the first place. Nonthermal plasmas and electric fields have been studied in the past for changing the behavior of flames, for activating fuels, oxidants and/or fuel-oxidant mixtures, and converting (into H2 and CO) of fuel-air mixtures (Yagodnikov et al., 1994; Bromberg et al., 1998; Rosocha et al., 2003). Nonthermal plasma has been studied for a long time, especially used as after-treatment control devices for reduction of air pollutants emitted from the exhaust of engines or emission sources. However, there are few studies focused on activating intake air for enhancing combustion efficiency and reducing emissions of toxic pollutants such as PAHs. The aims of this study are to use nonthermal plasma increase the free radicals of intake air, enhance the combustion efficiency and reduce emissions.

METHODS AND MATERIALS

Test Automobile, Dynamometer, and Plasma System

The gasoline automobile (with catalyst) used in this study was Mitsubishi Lancer 1.6L, the vehicle identification number HK-8402, with the following characteristics: four cylinders; four strokes; direct injection; water-cooled; bore and stroke of 95 mm (Dia.) × 105 mm; total displacement of 1597 mL; maximum torque of 169 Nm at 1,950 rpm and maximum horsepower of 40 kW at 2,600 rpm. The test engine was a four-cylinder carburetor with the ignition order of 1-3-4-2, bore and stroke of 77 × 80 mm2, swept volume of 1,490 cm3, power generation rate of 6.88 kW/1,500 rpm, and compression ratio of 9:1. Testing was conducted according to FTP-75 (Federal Test Procedure). The FTP-75 has been used for emission certification of light duty vehicles in the U.S. The engine was installed and operated on a dynamometer (Model FE 60-100-150S; Borghi & Saveri Corp, Bologna, Italy). Before the first experiment was conducted, the engine was adjusted at the laboratory of the manufacturer following the factory specifications such as injection timing, fuel consumption rate, exhaust emission, etc. Prior to each set of experiment, the system, including gas tank, fuel filling system, cylinder, and the manifold of inlet air and exhaust, was first cleaned, followed by a dirty-up procedure which was a sequence of 15 minutes engine running and 45 minutes stop, repeated 10 times. Furthermore, both the lubricating oil and the oil filter were changed and the fuel tank was emptied. The used plasma systems were from Ex power Ltd. Co., China. The plasma system was installed in the intake of gasoline engine. A series of experiments were conducted using plasma systems with different voltage ranged from 2400 to 3600 V. We used a high voltage probe (Model FLK-80K-40, Fluke Electronics, Canada) together with a multi-meter to measure the high voltage. The high voltage probe can scale the high voltage down 1000 times. The experimental setup is shown in Fig. 1.

Sample Collection

PAH samples of both the particulate-phase and gas-phase were collected by using a PAH sampling system at a temperature below 52°C in order to avoid desorption of PAH collected by cartridges. Particulate-phase PAHs were collected in glass-fiber filters. Before sampling, the filters were placed in an oven at 450°C for 8 hrs to burn off any organic compounds that might have been present. Finally, the cleaned filters were stored in a desiccator for at least 8 hrs to achieve moisture equilibrium before being weighed. After the experiments, the filters were brought back to the laboratory and put in a desiccator for 8 more hrs to remove the moisture. They were then weighed again to determine the net mass of particles collected. Gas-phase PAHs were collected in a three-stage glass cartridge containing a polyurethane foam (PUF) plug followed by XAD-16 resin. The glass cartridge was packed with 5.0 cm of XAD-16 resin sandwiched between a 2.5-cm upper PUF plug and a 2.5-cm bottom PUF plug. Silicone glue was used to seal and hold these two pieces of PUF to prevent resin from leaking out during the sampling and extraction processes. The new PUF/resin cartridge was cleaned up by Soxhlet extracting for one day each with distilled water, methanol, dichloromethane and finally n-hexane for a total of 4 days and then these PUF/resin cartridges were placed in a vacuum oven at 60°C for 2 hrs to dry and to evaporate the residual solvent in them. After drying, each PUF/resin cartridge was individually wrapped in hexane-washed aluminum foil and stored in a refrigerator at 4°C and transported in clean screw-capped jars with Teflon cap liners before sampling. Each glass fiber filter was transported to and from the field in a glass box, which was also wrapped with aluminum foil. In this study, each of the three-stage PUF/resin glass cartridges was analyzed. We found that less than 5% of total gas-phase PAHs (i.e., stage1 + stage 2 + stage 3) were collected by stage 3 suggesting the breaking through of gas-phase PAHs was negligible.
**PAH Analysis**

Each collected sample (including particulate and gaseous PAH samples) was extracted in a Soxhlet extractor with a mixed solvent (n-hexane and dichloromethane; vol/vol, 1:1; 500 mL each) for 24 hrs. The extract was then concentrated with nitrogen (N₂), cleaned up with sodium sulfate and reconcentrated to exactly 1.0 mL by N₂. The PAH contents were determined with a Hewlett-Packard (HP) gas chromatograph (GC) (HP 5890A; Hewlett-Packard, Wilmington, DE, USA), a mass selective detector (MSD) (HP 5972), and a computer workstation (Aspire C500; Acer, Taipei, Taiwan). This GC/MSD was equipped with a capillary column (HP Ultra 2, 50 m × 0.32 mm × 0.17 μm) and an automatic sampler (HP-7673A) and operated under the following conditions: injection volume of 1 μL; splitless injection at 310°C; ion source temperature at 310°C; oven temperature from 50 to 100°C at 20 °C/min, 100 to 290°C at 3 °C/min, and held at 290°C for 40 minutes. The mass of primary and secondary PAHs ions were determined by using the scan mode for pure PAH standards. The PAHs were qualified by using the selected ion monitoring (SIM) mode. The total-PAH data for the gasoline-engine exhaust were the summation of 21 individual PAHs.

The GC/MSD was calibrated with a diluted standard solution of 16 PAH compounds (PAH mixture-610M; Supelco, Bellefonte, PA, USA) plus five additional individual PAHs obtained from Merck (Darmstadt, Germany). Analysis of serial dilutions of PAHs standards showed the detection limit (DL) for GC/MSD was between 28 pg (pico-gram) and 324 pg for the 21 PAH compounds. Ten consecutive injections of a PAH 610-M standard yielded an average relative standard deviation of GC/MSD integration area of 6.96%, within a range of 4.33% to 9.76%. The R² of calibration in 21 PAH compounds ranged from 0.9945 (CYC)–0.9993 (PA). Following the same experimental procedures used for sample treatment, recovery efficiencies were determined by processing a solution containing known PAH concentrations. This study showed the recovery efficiencies for the 21 PAH compounds ranged from 0.881 to 0.952, with an average value of 0.918. Analyses of field
blanks, including aluminum foil, glass-fiber filters, and PUF/XAD-16 cartridges, revealed no significant contamination (GC/MSD integrated area < detection limit).

**Data Analysis**

The total PAH concentration was the sum of the concentrations for the 21 PAH compounds in each collected sample. To assess PAH homologues distribution for each collected sample, total PAHs were further classified into three categories of the LM-PAHs, MM-PAHs, and HM-PAHs. Moreover, considering that several PAH compounds are known human carcinogens, the carcinogenic potencies of PAH emissions from each emission source were also determined. In principle, the carcinogenic potency of a given PAH compound is assessed on the basis of its benzo[a]pyrene equivalent concentration (BaPeq). Calculation of the BaPeq concentration for a given PAH compound uses its toxic equivalent factor (TEF), which represents the relative carcinogenic potency of the given PAH compound, using benzo[a]pyrene as a reference compound to adjust its original concentration. Among the list of TEFs available (Chu and Chen, 1984; Thorslund and Farrer, 1991; Nisbet and LaGoy, 1992; Petry et al., 1996), the one by Nisbet and LaGoy (Nisbet and LaGoy, 1992) has been demonstrated to best reflect the actual state of knowledge of the toxic potency of each individual PAH species. Based on this TEF, the carcinogenic potency of total PAHs (total BaPeq) can be assessed by the sum of BaPeq concentrations estimated for each PAH compound with a TEF.

**RESULTS AND DISCUSSION**

**Emissions of Traditional Pollutants**

A series of experiments were conducted using plasma-enhanced system with different voltage ranged from 2400 to 3600 V. The emission factors of traditional air pollutants from gasoline-engine exhaust were presented in Table 1. The emissions of THC, CO, NOx, CO2, CH4, and NMHC for PE0 (base) were 146, 1390, 197, 198000, 10.9, and 135 mg/km, respectively. For PE2400, the reduction of THC, CO, NOx, CO2, CH4, and NMHC were 16.3%, 7.85%, 29.9%, 8.27%, 6.42%, and 12.5%, respectively, when compared with PE0. For PE2800, they were 35.1%, 16.8%, 67.5%, 11.8%, 7.34%, and 29.9%, respectively, when compared with PE0. For PE3200, they were 57.1%, 60.2%, 70.7%, 19.8%, 50.0%, and 57.7%, respectively. For PE3600, they were 57.8%, 61.0%, 71.5%, 20.5%, 51.3%, and 59.7%, respectively. The experimental results show that the reduction of pollutant emissions was enhanced with the increase of voltage. As mentioned before, plasma is typically an ionized gas, and is usually considered to be a distinct state of matter in contrast to gases because of its unique properties. In plasma state, the electrical conductivity is very high. Also, electrons, ions, and neutrals can be distinguished by the sign of their charge so that they behave independently and can interact simultaneously with many others in many circumstances such as liquids and gases. The reduction was attributed to the increase in free radicals and other reactive species in the intake air. Similar result was found (Warnatz et al., 1996). In addition, as shown in Table 1, the voltage had a significant effect on the reduction of pollutant emissions when the voltage was increased from 2400 to 3600 V. However, the voltage had a slight effect on the reduction of pollutant emissions when the voltage was increased from 3200 to 3600 V. Thus, the optimum voltage of plasma-enhanced system was 3200 V.

**Fuel Consumption**

Table 2 show the fuel consumptions of gasoline-engine exhaust under plasma-enhanced system with different voltage. The fuel consumptions of PE0, PE2400, PE2800, PE3200, and PE3600 were 72.8, 68.3, 67.3, 63.9, and 63.6 mL/km, respectively. The mean fuel savings were 6.11%, 7.52%, 12.1%, and 12.5% for PE2400, PE2800, PE3200, and PE3600, respectively, when compared with PE0 (72.8 mL/km). Fuel savings increased with increasing voltage. The above results indicate that plasma did increase free radicals and other reactive species in the intake air causing higher combustion efficiency and lower fuel consumption. A slight effect on the energy saving was found when the voltage was increased from 3200 to 3600 V. Therefore, the best performance was PE3200 with fuel saving ~12%.

**Emission of PM and SOF from the Exhaust of the Gasoline Engine**

The PM emission from the exhaust of the gasoline engine was shown in Fig. 2. Particulate matter from an engine includes three main components, soot formed during combustion, heavy hydrocarbon condensed or absorbed on the soot, and sulfates. The PM emission of PE2400, PE2800, PE3200, and PE3600 were all smaller than with PE0. The mean reduction fractions of PM from the exhaust of the gasoline engine were 16.5%, 30.2%, 42.8%, and 43.0% for PE2400, PE2800, PE3200, and PE3600, respectively (Fig. 2), when compared with PE0 (1.27 mg/m³). The result of PM reduction may be attributed to lower soot formation and soluble organic fraction (Akasaka et al., 1997; Kimura et al., 1999; Kimura et al., 2001). In engine
exhaust, the soluble organic fraction (SOF) is derived partly from the lubrication oil, partly from unburned fuel and partly from compounds during combustion. The SOF value was 52.8%, 43.6%, 35.2%, 27.4%, and 26.8% for PE0, PE2400, PE2800, PE3200, and PE3600, respectively. Conclusively, voltage had a significant effect on the pollutant reduction when the voltage was increased from 2400 to 3600 V but it had a slight effect on the PAH reduction when the voltage was increased from 3200 to 3600 V. The optimum voltage of plasma-enhanced system was 3200 V with best PAH reduction by 20.7% for total PAHs concentration and 20.5% for total BaPeq concentration from the gasoline exhaust followed a similar tendency. The mean reduction fractions of total BaPeq concentration from the gasoline exhaust were 10.9%, 14.4%, 20.5%, and 20.8% for PE2400, PE2800, PE3200, and PE3600, respectively, when compared with PE0 (3.27 μg/m³). The experimental results indicate that PAH emissions can be reduced due to free radicals and other reactive species in the intake air using plasma system. As shown in Table 2 and Table 3, the reduction tendency of traditional pollutants and PAHs were similar. Conclusively, fuel consumption increased with increasing voltage. The mean fuel savings were 6.11%, 7.52%, 12.1%, and 12.5% for PE2400, PE2800, PE3200, and PE3600, respectively, when compared with PE0 (240 μg/m³). LMW PAH concentrations in test fuels were all higher than MMW and HMW PAH. This result responded to previous experimental results: PAH content was primarily dominant in LMW PAHs (Mi et al., 1998; Mi et al., 2001). Total BaPeq emission concentration in the gasoline exhaust followed a similar tendency. The mean reduction fractions of total BaPeq concentration from the gasoline exhaust were 10.9%, 14.4%, 20.5%, and 20.8% for PE2400, PE2800, PE3200, and PE3600, respectively, when compared with PE0 (3.27 μg/m³). The experimental results indicate that PAH emissions can be reduced due to free radicals and other reactive species in the intake air using plasma system. As shown in Table 2 and Table 3, the reduction tendency of traditional pollutants and PAHs were similar. Conclusively, voltage had a significant effect on the pollutant reduction when the voltage was increased from 2400 to 3200 V but it had a slight effect on the PAH reduction when the voltage was increased from 3200 to 3600 V. The optimum voltage of plasma-enhanced system was 3200 V with best PAH reduction by 20.7% for total PAHs concentration and 20.5% for total BaPeq concentration. COMPARATIVE STUDIES OF CURRENT GASOLINE ENGINES USING PLASMA SYSTEMS. As shown in Table 2 and Table 3, the five predominant species of PE0 were Nap (34.1%), Ant (25.1%), Flu (15.6%), FL (5.50%), and Pyr (4.04%). The five predominant species of PE2400 were Nap (34.4%), Ant (25.1%), Flu (15.5%), FL (5.48%), and Pyr (4.01%). The five predominant species of PE2800 were Nap (34.4%), Ant (25.1%), Flu (15.5%), FL (5.42%), and Pyr (3.99%). The five predominant species of PE3200 were Nap (33.7%), Ant (25.4%), Flu (15.6%), FL (5.56%), and Pyr (4.03%). The five predominant species of PE3600 were Nap (34.0%), Ant (25.1%), Flu (15.8%), FL (5.46%), and Pyr (3.98%). The results indicate that the five predominant species of PE0, PE2800, PE320, and PE3600 were similar, comprising of Nap (33.7%–34.4%), Ant (25.1%–25.4%), Flu (15.5%–15.8%), FL (5.42%–5.56%), and Pyr (3.98%–4.04%). Conclusively, Nap, Ant, Flu, FL, and Pyr account for~80% of total PAH emissions, although the fractions of individual species varied in each test fuel.

CONCLUSIONS

Plasma activation is an innovative concept for converting liquid or gaseous fuels into reactive species. It involves precracking the fuel into the requisite reactive species by the application of atmospheric-pressure plasmas. However, there are few studies focused on activating air for enhancing combustion efficiency and reducing emissions of toxic pollutants such as PAHs. In this study, a plasma was used to increase the free radicals of intake air and enhance the combustion efficiency. Experimental results show that the reduction of pollutant emissions was enhanced with the increase of voltage. The mean fuel savings were 6.11%, 7.52%, 12.1%, and 12.5% for PE2400, PE2800, PE3200, and PE3600, respectively, when compared with PE0 (72.8 mL/km). Fuel savings increased with increasing voltage. The above results indicate that plasma did increase free radicals and other reactive species in the intake air causing higher combustion efficiency and lower fuel consumption. The mean reduction fractions of total PAH concentration

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**Table 2. Fuel consumption of gasoline engine.**

<table>
<thead>
<tr>
<th></th>
<th>Fuel consumption (mL/km)</th>
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<tbody>
<tr>
<td>PE0</td>
<td>72.8</td>
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<tr>
<td>PE2400</td>
<td>68.3</td>
</tr>
<tr>
<td>PE2800</td>
<td>67.3</td>
</tr>
<tr>
<td>PE3200</td>
<td>63.9</td>
</tr>
<tr>
<td>PE3600</td>
<td>63.6</td>
</tr>
</tbody>
</table>

**Fig. 2. PM emission in the exhaust of the gasoline engine.**

**Fig. 3. SOF in the exhaust of the gasoline engine.**
Table 3. PAH concentration in the exhaust of diesel generator.

<table>
<thead>
<tr>
<th>PAH</th>
<th>TEFa</th>
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</thead>
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<tr>
<td></td>
<td>PE0</td>
</tr>
<tr>
<td>Nap</td>
<td>81.6</td>
</tr>
<tr>
<td>AcPy</td>
<td>4.85</td>
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<tr>
<td>Acp</td>
<td>0.271</td>
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<tr>
<td>Flu</td>
<td>37.4</td>
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<tr>
<td>PA</td>
<td>8.88</td>
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<tr>
<td>Ant</td>
<td>60.1</td>
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<tr>
<td>FL</td>
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<tr>
<td>Pmr</td>
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<tr>
<td>CYC</td>
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<td>BaA</td>
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<td>CHR</td>
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<td>BbF</td>
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<tr>
<td>BKF</td>
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<tr>
<td>BeP</td>
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<td>BaP</td>
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<tr>
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<tr>
<td>DBA</td>
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<tr>
<td>BbC</td>
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<td>COR</td>
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</table>

ΣLMW-PAHs 193 173 166 153 152 -
ΣMMW-PAHs 30.2 26.9 25.6 24.0 23.6 -
ΣHMW-PAHs 16.2 14.4 13.9 12.8 12.8 -
Total PAHs 240 214 206 190 189 -
Total BaPeq 3.27 2.91 2.80 2.60 2.59 -

a Nisbet and LaGoy, 1992.
b No TEF has been suggested.

from the gasoline exhaust were 10.6%, 14.2%, 20.7%, and 21.2% for PE2400, PE2800, PE3200 and PE3600, respectively, when compared with PE0 (240 μg/m³). The mean reduction fractions of total BaPeq concentration from the gasoline exhaust were 10.9%, 14.4%, 20.5%, and 20.8% for PE2400, PE2800, PE3200 and PE3600, respectively, when compared with PE0 (3.27 μg/m³). Conclusively, the reductions of pollutant emissions increased with increasing voltage and energy saving increased with increasing voltage as well. The optimum voltage of plasma-enhanced system was 3200 V with reductions of THC by 57.1%, CO by 60.2%, NOx by 70.7%, CO2 by 19.8%, CH4 by 50.0%, NMHC by 57.7%, PM by 42.8%, and total PAHs by 20.7%, and total BaPeq by 20.5%, and fuel savings by 12.1%. However, high variability is usually associated with emissions from different motor vehicles. Further investigations on more vehicles are needed in the further.

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