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Removal of emission gas COx, NOx and SOx from automobile using non-thermal plasma

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Abstract. Non-polluted plasma muffler prototype has done in fuel engine vehicle. Pilot scale improvement has done by integrating reduction system into vehicle muffler from previous position outside the muffler. High voltage that used to develop plasma condition comes from 12V34Ah accumulator which connected with electronic equipment and it can be able to develop voltage up to 20kV. Exhaust gases reduction ability has done by varying engine rotation. Plasma muffler appearance in vehicle doesn't change outside dimension of its original muffler and it reactor placement in muffler has a function to change resonator chamber function and make this muffler still fulfil muffler standardization with more performance in reducing exhaust gases (CO_x, NO_x, HC) . Optimal reduction level made at 2200 rpm for CO_x is 86.52%, for CO is 88.93%, for HC is 97.34% and for NOx at 4600 rpm is 76,19%. The residue formed in the muffler has been tested with FTIR. Of all possible feeds, the compound constituent of the NOx reduction product is ammonium carbonate ((NH₄) 2CO₃), ammonium sulphite ((NH₄)2SO₃), and also ammonium sulphate ((NH₄) 2SO₄).

Keywords: Non-Thermal Plasma, gas emission, motorized vehicle, plasma, reduction.

1. Introduction

Pollution of greenhouse gas emissions has been widely contaminated to our environment. In the case of municipal waste, these greenhouse emissions direct and indirect burden the environment In the case of municipal waste, these emissions direct and indirect burden the environment among others methane (CH_4) , carbon dioxide (CO_2) , ammonia (NH_3) , nitrate (NO) and nitrous oxide (N_2O) [1]. In addition, emissions from motorized vehicles also greatly affect the cleanliness of the air in the city and result in a greenhouse effect for the earth, the greenhouse gas emissions for road transport sector in Mumbai Metropolitan Region (MMR) using fuel consume [2] emissions CO₂ by automobiles in Brazil [3]. Nonthermal plasma technology in recent years has been tried to solve these environmental pollution [4-6] pulsed discharge plasma [7]. Efforts to reduce greenhouse gas emissions generated by motorized vehicles have been widely used using filters. Some researchers have also tried to realize gas emission reduction systems from motor vehicles by using non thermal plasma especially corona discharge [8]. The use of hybrid methods of combining non-thermal plasma and gas circulation systems has been tried to reduce NO_x. This system does not require precious metal catalysts or hazardous chemicals such as urea and ammonia. In this system, NO_x in diesel emissions is treated by adsorption and desorption by



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adsorbents and reduction of non-thermal plasma [9]. Non-thermal plasma technology (NTP) has also been tried to be applied in the production of diesel engine exhaust emissions along with using a dielectric barrier (DBD) reactor, the particles released by diesel combustion can be reduced [10]. This paper reports on the results of research on the reduction of emissions of gases released by diesel engine cars in a stationary state. The plasma technology used is non-thermal plasma (NTP) which is generated by corona discharge.

2. Methods

Test-fuelled motorized vehicles. Gas Analyser (Horiba PG-250) was used to measure the concentration of emission gases (CO_x , SO_x , NO_x , and HC) before and after. The reduction process, the CO_x , NO_x and HC reducing systems were strung together as shown in Fig. 1. Plasma generator is carried out by connecting the high voltage cable from the high voltage provider to the wire electrode and the cylinder (field) electrode shaft connected to ground. Then the multimeter is installed in series on the circuit to measure the input current.

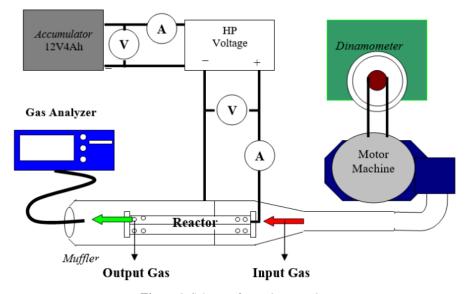


Figure 1. Scheme of experiment series.

After reducing in the reactor there is an aerosol-shaped crust, and the crust is analysed by FTIR Spectroscopy (Fourier Transformation Infrared, Shimadzu Hyper FTIR-820 IPC). Current pulse was detected with a Tektronix oscilloscope (model 7633).

Reduction of CO_x , NO_x and HC after the reactor is cleaned from impurity particles that may be present in the reactor. Vehicle gas emissions from the muffler are measured using a gas analyser to determine the concentration of CO_x NO_x and initial HC (concentration of CO_x NO_x and HC before reduction) as well as to ensure that the reduction of CO_x NO_x and HC only occurs due to the corona incandescent plasma discharge in the reactor. After the gas is in the reactor, the voltage from the high voltage supply system is given to the reactor by connecting it with a dc voltage source in the form of a 12V34 Ah accumulator. Gas emissions of vehicles in the reactor are treated with plasma conditions in a continuous flow state. This is intended to be able to observe the condition of the gas in the plasma if the gas is in a flowing state. The reduction is given by varying the motor rotation by changing the engine and gear rotation. After the reduction process is carried out then the gas is removed from the reactor to measure CO_x and HC concentrations using a gas analyser. CO_x and HC concentrations that are read are

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 CO_x and HC concentrations after being reduced. The percentage of reduction in CO_x , NO_x and HC can be determined using the following equation. Reduction Percentage Emission Gas (EG) =

 $\left(1 - \frac{C_{tEG}}{C_{oEG}}\right) \times 100\%$ with C_{oEG} is concentration emission gas before treatment, C_{tEG} is concentration

emission gas after treatment. In this research, the determination of the corona plasma discharge area was initially carried out by looking for the characteristics of the plasma reactor by slowly applying the voltage to the plasma reactor and recording the current increase readable in the multi-tester in conditions with and without flue gas flow. Before reducing, the vehicle exhaust gas is put into the plasma reactor to measure the concentration of pollutants CO_2 , CO and HC by using a gas analyser. The measured concentrations of CO_2 , CO and HC before reducing this is a reference in comparing the levels of CO_2 , CO and HC plasma reactor reduction.

3. Results and Discussion

3.1. Reducing Pollutants CO_x, NO_x and HC

In the condition of corona incandescent plasma discharge, the chain ionization that occurs causes the dissociation of CO_2 , CO and HC gases and other gases contained in flue gases such as water vapour (H₂O) and HC into ions, electrons, and energetic radicals. The process of forming radicals by multiplying electrons is called the initiation stage in the mechanism of free radical reaction. Next at the propagation stage in the mechanism of the free radical reaction, unstable energetic radicals will react with the flue gas molecules contained in the plasma reactor prototype so that new dissociations occur from the gas molecules.

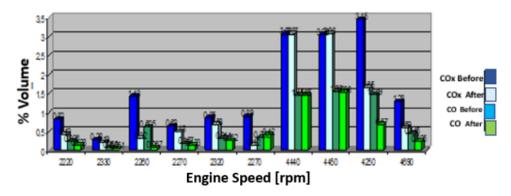


Figure 2: Reducing CO_x, CO in a car muffler with a modified plasma reactor in it and connected to a high voltage source.

According [8] dissociation in CO₂ will produce CO, C*, and O*, the dissociation in CO will produce C* and O*, while dissociation in water vapour (H₂O) will produce H* and OH*. The mechanism of free radical reaction ends when the radicals produced from the dissociation process react to form a new compound that is stable so that the concentration of CO₂ and CO in the exhaust gas becomes reduced. The reduction ability with the generation of corona discharge in the plasma reactor mounted on the muffler can be seen in the reducing graph can be seen in Fig. 2.

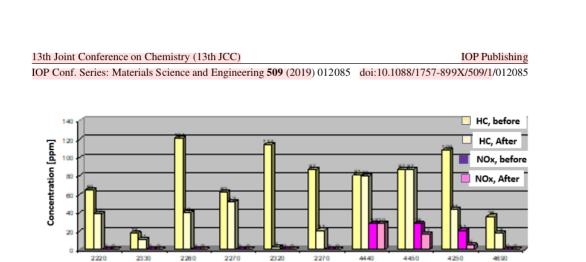


Figure 3. Reduction of NO_x , HC in car mufflers with modified plasma reactors in it and connected to high voltage sources.

Engine Speed [rpm]

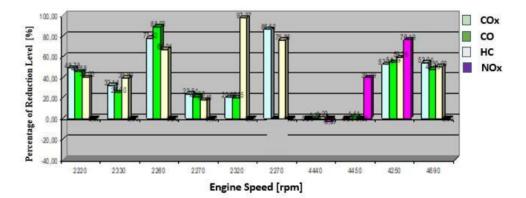


Figure 4. Reduction level of CO_x , CO, NO_x , HC in car mufflers with modified plasma reactors in it and connected to high voltage sources

The optimum reduction rate for CO_x gas is obtained at 2200 rpm which is 86.52%, CO at 2200 rpm is 88.93%. In our research, we obtained NO_x released by diesel cars which could be reduced by 76.19%. Hydrocarbons (HC) emitted by diesel engine exhaust with non-thermal plasma (NTP), plasma corona type can be eliminated by 97.37%. This research was conducted with the condition of stationary diesel engine with a rotation of 4600 rpm. The results obtained in this study show similarities with the results obtained [11]. Reduction of NO_x gas uses non-thermal plasma (NTP) and exhaust gas recirculation. These researchers also used pilot scale systems on stationary diesel engines. In this study, about 57% of NO_x from exhaust can be reduced continuously for 58 hours.

3.2. Analysis of solids formed in the reactor

In this study we have analysed the reaction products that occur in the reactor. The product was in the form of dust. The analysis was done using FTIR. The results of FTIR analysis show that there has been a free-radical reaction in the reactor on plasma state. It was evidenced in the reaction product analysis. Fig. 5 shows the FTIR absorption spectrum of residual (reaction product) from the gas emission reduction of a four-wheeled motor vehicle with gasoline fuel (Fig. 5). In this spectrum the presence of hydrocarbon compounds in the combustion process is indicated by the presence of absorption bands of hydrocarbon groups such as $-CH_2^-$. While the identification of NH groups such as the formation of -NH-groups and $-NH_2$ groups in the medium absorption bands 1541.0 cm⁻¹ and 1624.0 cm⁻¹. We found also

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stretching -NH₂ groups on 3425.3 cm⁻¹. The absorption band very prominent which shows the possibility of formation of NH₄⁺ cation as a result of the reaction between N*, that produced from NO_x or N₂. We detected also with H* and H⁺ produced from H₂O. The observation of the functional groups -CH₂⁻, CH₃-CO-O, CH₃-C, CO₃^{-2,}, -NH⁻, -NH₂, and -NH₂ stretching, shows that during the plasma condition in the reactor, the free radicals reactions and recombination between compounds have occurred. contained in the exhaust gas of the motorized vehicle tested. This gives an explanation that NO_x gas, water vapour (H₂O), HC and other compounds such as CO, CO₂ and SO₂ in the reactor after the generation of corona incandescent plasma, is no longer in a gas state but has turned into an ion or radical and react with each other, thus forming new compounds which include crust which settles in the reactor, so that NO_x concentration decreases.

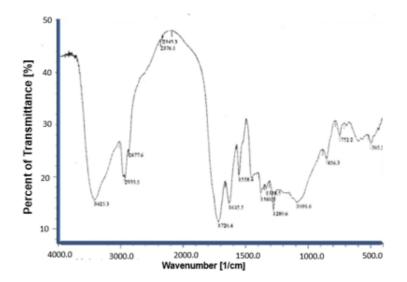


Figure 5. IR Spectrum of reaction products in the form of dust in side of reactor.

The formation of NH_4^+ cations can occur through the hypothesis of a gradual reaction between N* radicals and H* radicals and H⁺ ions as follows:

$$N^* + H^* \longrightarrow NH^*$$
 (R.1)

$$H^* + NH^* \longrightarrow NH_2^*$$
 (R.2)

$$H^* + NH_2^* \longrightarrow NH_3$$
 (R.3)

$$H^+ + NH_3 \longrightarrow NH_4^+$$
 (R.4)

The radical N* in the NH₄ cation is obtained from NO_x dissociation. The dissociation process can be explained by the following reaction hypothesis

$$e + NO_2 \longrightarrow NO + O^* + e$$
 (R.5)

$$NO + O^* \longrightarrow N^* + O_2 \tag{R.6}$$

The radical O* formed from dissociation of CO_2 , CO, NO_2 , NO, or H_2O can bind to CO_2 and CO to form $CO_3^{2^2}$, the process can be explained by the following reaction hypothesis:

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$$O^* + CO \longrightarrow CO_2$$
 (R.7)

$$O^* + CO_2 \longrightarrow CO_3^2$$
 (R.8)

While the formation of SO_3^{3-} and SO_4^{2-} can occur through a gradual reaction between O* radicals formed from dissociation of CO_2 , CO, NO_2 , NO, or H_2O related to SO or SO_2 which is explained by the following reaction:

$$O^* + SO \longrightarrow SO_2$$
 (R.9)

$$O^* + SO_2 \longrightarrow SO_3^{2-}$$
 (R.10)

$$O^* + SO_3 \longrightarrow SO_4^{2-}$$
 (R.11)

Then the formed anion compound can react with the NH₄⁺ cation through the reaction:

$$2NH_4^+ + CO_3^{2-} \longrightarrow (NH_4)_2CO_3 \tag{R.12}$$

$$3NH_4^+ + SO_3^{2-} \longrightarrow (NH_4)_2 SO_3$$
 (R.13)

$$2NH_4^+ + SO_4^{2-} \longrightarrow (NH_4)_2SO_4 \tag{R.14}$$

From the discussion above, it can be proposed that the gas emission reducing from auto mobile using a corona incandescent plasma reactor, the crustal compounds (reaction products) are ammonium carbonate ((NH4)2CO₃), ammonium sulphite ((NH₄)2SO₃), and ammonium sulphate ((NH₄)2SO₄). According to [10, 11] reducing CO_x and SO_x will be effective if NO_x has been reduced first and will be easier to clean from the exhaust pipe of motor vehicle exhaust due to the large particle diameter. This is also evident in this study because nitrogen is a constituent of ammonium which also functions as a binder of CO_x and SO_x in vehicle exhaust gases.

4. Conclusion

Emission reduction system for plasma technology can be installed on motor vehicle exhausts and does not change vehicle performance. Radical reactions that occur from emission gases in plasma conditions can take place in the absence of added additives such as NH_3 , Ar and H_2O steam. The optimum reduction rate for CO_x gas is obtained at 2200 rpm which is 86.52%, CO at 2200 rpm is 88.93%, HC at 2200 rpm is 97.37% and NO_x at 4600 rpm is 76.19%. This research can be continued on basic research so that it can be revealed the mechanism of the reaction that occurs in plasma conditions so that the gas emissions are reduced and the presence of aerosols is formed. This basic study is expected to produce a new theory about the mechanism of radical reactions that occur in plasma conditions

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 PAGE 1
 PAGE 2

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 PAGE 3

 PAGE 4
 PAGE 5

 PAGE 5
 PAGE 6

 PAGE 7
 PAGE 8