# NOx abatement using in-situ fuel-based reductant under rotating arc plasma condition

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**Abstract:** In this study, NOx reduction using hydrocarbon as reductant which was reformed by rotating arc plasma was carried out. The fuel itself was utilized as hydrocarbon source. To enhance the reducibility, rotating arc plasma reformed the fuel to hydrogen, light hydrocarbon, oxygenated hydrocarbon, and so on. The effects of carbon to oxygen ratio and additional ozone injection over the temperatures from room temperature to 300°C was investigated

Keywords: Plasma, Hydrocarbon selective catalytic reduction, De-NOx, Ozone

### 1. Introduction

Nitrogen oxides (NOx) emissions are the major cause of human health and environmental problems owing to the contribution of acid rain [1-2]. Technologies have been developed to reduce the NOx emissions for meeting the stringent emissions regulations through engine design optimization, internal modification including low temperature combustion or wet methods, after-treatment technology including three-way catalyst, selective catalytic reduction or plasma technology. Among these technologies, selective catalytic reduction (SCR) can efficiently reduce NOx to form  $N_2$  by the addition of urea injection. Its operation temperature requires above 250°C for complete [3-4]. removal of NOx Furthermore, periodic replenishment of urea solution as a reductant is necessary for catalytic reduction, which leads to necessity of additional space for reductant. Consequently, the system become complex and expensive due to urea injection [5-6]. In order to overcome these limits for utilizing diesel vehicles, hydrocarbon-SCR (HC-SCR) can be applied to simplify the system. Among the catalysts studied over the years, one of the promising alternatives to Cu/ZSM-5 which is mostly pronounced for high temperature applications is Ag/Al<sub>2</sub>O<sub>3</sub> due to its excellent activity, good selectivity to N<sub>2</sub> and tolerance to SO<sub>2</sub>. However, most of the studies done so far utilized addition of hydrogen or light hydrocarbons, not fuel itself. There is a need of diesel fuel itself as hydrocarbon reductant source of HC-SCR, which gives simplicity of the system. Plasma assisted HC-SCR system has been attractive alternative for low temperature NOx removal due to its on-board capability of flexible reactive products including hydrogen, light HCs and oxygenated hydrocarbons (OHCs) from diesel itself as reductants. Main purpose of the technology is to reduce NOx species into N<sub>2</sub> after the catalyst at lower temperature through generated active species by means of plasma driven HC reforming process which is being injected to the NOx stream.

## 2. Experimental

This study was performed initially in rotating arc plasma reaction system that was proposed for reforming HC to OHC, light hydrogen, etc. Then, this reaction system was coupled to the test bench of HC-SCR system for further investigation of De-NOx performance. The experimental setup for a rotating arc plasma reforming system consisted of a rotating arc plasma reactor, a power supply system, measurement system, gas flow controllers, fuel syringe pump, cold trap as shown in Fig. 1. Fig. 2 illustrates a schematic diagram of the plasma assisted HC-SCR test bench replicating heavy duty diesel engine exhaust flow conditions. Reformer gas from the rotating arc plasma reactor was supplied to the HC-SCR system through a mixing chamber.



Fig. 1. Schematic of the experimental setup for rotating arc plasma system.



# 3. Result and discussion

Flexible production of rotating arc plasma driven by reformer gas was investigated along with carbon to oxygen ratio from fuel lean as 0.7 to rich condition as 2.1 with increment of 0.35 at constant SEI of 900 J/l. Carbon balance decreased significantly when C/O<sub>2</sub> ratio reached 2.1 due to the lack of oxygen. Carbon dioxide (CO<sub>2</sub>) was main product besides small amount of carbon monoxide

(CO) and hydrogen at C/O<sub>2</sub> of 0.7 where almost complete combustion occurred due to near stoichiometric condition as fuel/air equivalence ratio of 1.15. As the fuel injection rate increased further, partial oxidation and hydrocarbon cracking induced the elevation of CO and H<sub>2</sub> concentration including light hydrocarbons (C1&C2). Then, CO and H<sub>2</sub> tended to decrease at C/O<sub>2</sub> of 2.1. This is because oxygen concentration is not enough to conduct partial oxidation at that condition, which leads to increased reaction tendency to hydrocarbon cracking.

The concentration of liquid product with respect to  $C/O_2$  ratio was also measured. Noticeable products detected were methanol and acetic acid of which concentration increased as the increase of  $C/O_2$ . Small amount of ethanol and benzaldehyde were observed and did not show significant difference throughout the conditions. Dodecane was not detected in the liquid products. It was found that liquid product concentration increased as the rise of  $C/O_2$  ratio.

Comparison of De-NOx performance to realize the effect of ozone addition was experimentally investigated on HC-SCR test bench with total exhaust gas flow rate of 500 lpm. The De-NOx performance along with the catalyst temperature (T<sub>cat</sub>) from room temperature to 300°C in the absence of ozone was observed. The room temperature experiment was conducted for the reference to compare with ozone condition. There was not much difference until the T<sub>cat</sub> reached 100°C. However above 100°C, NOx removal tended to improve at C/O<sub>2</sub> ratio of 1.4 and 1.75. For higher C/O<sub>2</sub> ratio of 2.1, NOx conversion became slightly lower due to high amount coke generated during reforming process which is detrimental to the catalyst. In the presence of additional ozone, NOx conversion was enhanced at low catalyst temperatures of room temperature and 100°C for lower C/O<sub>2</sub> ratios as 0.7 and 1.05. However, it is obvious that there is small amount of reductant at these conditions. It was thought that this happened due to NO oxidation to NO<sub>2</sub> in the presence of hydrogen, which leads to the adsorption of NOx on Ag catalyst surface. Also, there could be undetected NOx products such as  $N_2O_5$ . There was substantial increase of NOx removal at T<sub>cat</sub> of 250°C where NOx conversion was near 60% for C/O<sub>2</sub> ratios of 1.05, 1.4 and 1.75.

### 4. Conclusion

In this study, the effects of carbon to oxygen ratio on fuel-based reductant generation using rotating arc plasma was investigated for NOx removal. At first, the gas and liquid products from the plasma reformer without ozone was analyzed. Furthermore, the case of ozone addition on downstream of the reformer was compared to the case without ozone. Finally, the plasma driven hydrocarbon products were introduced to the SCR system that was carried out in 500 lpm bench scale system without external reductant. In the bench scale system, over 95% of NOx conversion was shown below 300°C. In addition, it was found that the proportion of liquid product such as alcohol, acid could increase when the external ozone was fed. It was confirmed that low temperature De-NOx performance could be enhanced by these liquid products, especially ethanol. By using fuel-based reductant from plasma reactor, NOx could start to be reduced even at 100°C.

### **5.References**

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