

Chemical, Petroleum and Environmental Engineering

Experimental Investigation for the Removal of Toxic Gases from Vehicle Exhaust using Non-Thermal Plasma

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ABSTRACT

In this study, the use of non-thermal plasma theory to remove toxic gases emitted from a vehicle was experimentally investigated. A non-thermal plasma reactor was constructed in the form of a cylindrical tube made of Pyrex glass. Two stainless steel rods were placed inside the tube to generate electric discharge and plasma condition, by connecting with a high voltage power supply (up to 40 kV). The reactor was used to remove the contaminants of a 1.25-liter 4-cylinder engine at ambient conditions. Several tests have been carried out for a ranging speed from 750 to 4,500 rpm of the engine and varying voltages from 0 to 32 kV. The gases entering the reactor were examined by a gas analyzer and the gases concentration ratio are recorded in the inlet of the reactor and after they are released from the reactor after the chemical processes associated with the electric discharge applied to the gases inside the reactor. As a final result, convergent removal rates of gases were obtained but under different conditions, the best ratios were: NO_x 72.32% (at a rotational speed of 3500 rpm and an applied voltage of 25kV, HC 69.46% (at 1500 rpm and 30kV, CO 66.66% (at 3000 rpm and a range of voltage from 25 to 32kV) and CO₂ 72.44% (at 3500 rpm and 27 kV).

Key Words: nonthermal plasma (NTP), NO_x, CO, CO₂, HC.

التحقيق العملي لإزالة الغازات السامة من عوادم المركبات باستخدام البلازما غير الحرارية

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مدرس

الهندسة الكهروميكانيكية – الجامعة التكنولوجية

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طالب ماجستير

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الخلاصة

في هذه الدراسة تم التحقق عمليا من استخدام نظرية البلازما غير الحرارية لإزالة الغازات السامة المنبعثة من المركبات. تم بناء مفاعل يستخدم نظرية البلازما غير الحرارية على شكل انبوبة اسطوانية الشكل مصنوعة من مادة الزجاج الحراري (بايركس). تم وضع قضيبين من مادة الفولاذ المقاوم للصدأ داخل الانبوبة للحصول على التفريغ الكهربائي وحالة البلازما

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وذلك بربطهم بمجهاز فولطية عالي يصل الى 40 كيلو فولط. تم استخدام هذا المفاعل للتخلص من ملوثات محرك سيارة سعة 1.25 لتر رباعي الاسطوانات وذلك عند الظروف المحيطة. تم إجراء تجارب عديدة حيث كانت سرعة المحرك تتراوح ما بين 750 الى 4500 دورة بالدقيقة وبفولطيات مختلفة من صفر إلى 32 كيلو فولط. تم فحص الغازات الداخلة الى المفاعل عن طريق جهاز تحليل الغازات ومعرفة نسب هذه الغازات عند دخول المفاعل وعند خروجها بعدما تتم عليها العمليات الكيميائية المصاحبة للتفريغ الكهربائي المسلط على الغازات داخل المفاعل. كنتائج نهائية تم الحصول على نسب إزالة متقاربة للغازات ولكن بظروف مختلفة فكانت أفضل النسب هي كالتالي : أكاسيد النتروجين 72.32% (وذلك عند سرعة دوران محرك 3500 دورة بالدقيقة، وعند فولطية مسلطة 25 كيلو فولط)، الهيدروكربونات 69.46% (وذلك عند سرعة دوران محرك 1500 دورة بالدقيقة، وعند فولطية مسلطة 30 كيلو فولط)، أول أكسيد الكربون 66.66% (وذلك عند سرعة دوران محرك 3000 دورة بالدقيقة، وعند معدل فولطية مسلطة من 25 إلى 32 كيلو فولط) و ثاني أكسيد الكربون 72.44% (وذلك عند سرعة دوران محرك 3500 دورة بالدقيقة، وعند فولطية مسلطة 27 كيلو فولط).

الكلمات الرئيسية: البلازما غير الحرارية، أكاسيد النتروجين، أول أكسيد الكربون، ثنائي اكسيد الكربون، الهيدروكربونات

1. INTRODUCTION

1.1 Air Pollution

Air pollution is considered as one of the most dangerous and the biggest problems facing the entire world. It is the mean of the presence of any gaseous, liquid or solid in the ambient air with quantities that lead up to a damage in the (physiological and economic vitality) of plants, animals, humans, equipment and machinery, or affect the nature of objects The main sources of air pollution are combustion processes that are used in power plants, cars and other modes of transport, such as trains, ships, and airplanes, **Skalska, et al., 2010**. Moreover, there are also industrial processes such as oil refineries, petrochemical and other industries leading to increased air pollution, **Zare and Anvaripour, 2014**. These contaminants can be divided into primary pollutants that result from a particular process, such as smoke emitted from cars and secondary pollutants that are generated due to the reaction of natural air with primary pollutants, **Sharma, 2014**. The environment in Iraq suffers from many problems, their causes are back to the natural and human factors, wrong policies that isolated Iraq from the world for many years as a result of successive wars, the various international sanctions, and economic blockade. All these and other factors cause great pressure on the environment. The deterioration of air quality in Iraq had a negative impact on the environment and health situation where the rates of incidence of chronic respiratory diseases and allergies increased. The deterioration of air quality can be attributed to:

- 1.The significant increase in the number of vehicles in general, where old vehicles constitute a large proportion as well as the use of fuel that does not meet environmental standards.
- 2.The lack of national power generation due to the outdated stations and rely on home generators small and large that supply the houses located in the street, this process is accompanied by the emission of noise and pollutants.
- 3.Acts of sabotage in the transport line of oil and its derivatives, and the outbreak of the fires.
- 4.Citizen's asylum to waste incineration as an alternative to the weakness of the process of waste collection.
- 5.Over-harvesting of trees in general and palm particularly to cover the needs of the fuel for citizen which reduces the green spaces, **Ministry of Environment – Iraq, 2017**.

1.2 Pollution Control

The control of pollution is a process that reduces or eliminates the release of pollutants into the environment. It has been organized by different environmental agencies which set limitations on the discharge of pollutants into the (air, water, and land). It has been developing a various collection of systems and appliances to control the air, water, and solid waste pollution, **Redha, 2012**. There are many ways and methods available to remove unwanted polluting gases, such as scrubbers, adsorption, catalytic converter and other of traditional methods.



The process of using nonthermal plasma (NTP) technology to control the environment from pollution had been extensively studied because it is a promising and effective technology to control the pollution with higher energy efficiency. Through the use of non-thermal plasma, many toxic molecules can be treated and can be used in places that are difficult to use the traditional methods, **Penetrante, et al., 1993**.

In non-thermal plasma, the gas temperature is too close to the ambient temperature, but electrons are with high temperature, and as a result, a reaction through ionization is obtained, excitation and dissociation processes, **Alkawareek, et al., 2012**.

NTP can be generated through various types of gas discharge including DC glow discharge, radio frequency discharge, (DBD) dielectric barrier discharge, surface discharge, atmospheric pressure discharge, microwave discharge, and (CD) corona discharge, **Ming-Wei, et al., 2004**.

1.3 Researches of NTP Technology in Iraq

The use of non-thermal plasma in Iraq has taken place in some application such; generation of plasma needle as a type of non-thermal plasma for the purpose of antimicrobial activity, effect of non-thermal plasma on blood coagulation and for water pollution removal, **Kadhim and Heba, 2016**, some researches about the treatment of Bacteria with NTP, **Thamir, et al., 2015**, design an ozone generator with dielectric barrier discharge method, **Hamid and Hadeel, 2014**, and there is not a research or application found on the use of NTP for pollution control. This research is the first one in Iraq to study this technique with details and to be used for the removal of toxic gases that emitted every day.

2. EXPERIMENTAL WORK

The experimental apparatus used in this study is represented as a schematic diagram shown in **Fig. 1** and a real photograph of the system shown in **Fig. 2**. Toxic gases generated by the 1248cc engine are analyzed with NHA-506EN Gas Analyzer, which can measure the emission NO_x, CO, CO₂, and HC. **Fig.3 (a and b)** shows a cross-section view and a real photograph of the main section in the experimental work. Plasma reactor is a cylindrical tube made of Pyrex glass with an outer diameter of 70mm, a length of 300mm, a thickness of 2mm with 2 circular Teflon stoppers to prevent the gas from going out the tube and to bear the high temperature of the gas. The tube consists of one inlet pipe (with od of 12mm) for inlet gas, one exit pipe (with od of 12mm) for exit gas, one optional pipe, two electrodes (stainless steel rods with diameter of 6mm) is set inside the tube, reactor have two holes in every Teflon stopper these holes to set the electrodes in horizontal shape these holes were closed with silicone sealant from the outside for no gas leakage. The reactor receives the gas from the engine by stainless steel hose and exposed to the process inside the tube. Plasma reactor is set to operate with DC high voltage supply (0~40kv) connected with the electrodes that set in the reactor. Several samples are taken from the exhaust gas at different, operation condition and the rotational speed of car engine at the inlet of the reactor and the exit of it after process done.

3. RESULTS AND DISCUSSION

3.1 Exhaust Emission Characterization of the Engine

The gaseous emission has been measured before studying the effect of non-thermal plasma reactor on engine exhaust gases, at different engine rotated speeds and two types of engine loads namely (with & without load (25%)) as turning on the air-conditioning system as a load on the engine. In general, the concentration of NO_x varies from 20 ppm to 435 ppm, the CO augments from 0.00 %v.v to 0.07 %v.v, the total hydrocarbons (HC) concentrations vary from 20 ppm to 131 ppm and the range of CO₂ concentrations is from 2.66 %v.v to 15 % v.v.



3.1.1 Nitrogen Oxides (NO_x)

The NO_x concentration results are very complicated, it depends on combustion temperature, availability of oxygen, and time for the combustion process. As can be seen in **Fig. 4** the concentration of NO_x increases with increasing engine speed (output power) at constant load due to the increase in the cylinder temperature. This is due to the higher temperature caused by the better combustion process, so the maximum level of NO_x emission is obtained at maximum speed. At load condition, the concentration of NO_x was higher than the values at no loads this is due to better combustion process that leads to higher combustion temperature which favors NO_x formation. It has shown a sudden increase in the concentration of NO_x at the range between (2500 to 3000) rpm and then sharp decrease that may be due to some internal combustion processes.

3.1.2 The (unburned hydrocarbon) HC

The concentration of the hydrocarbons patterns is similar for the two engine conditions. As can be seen in **Fig. 5** after a drop in the concentration at 1500 rpm the concentration was changed slightly by changing the rotating speed of the engine. The very high engine speed reduces the volumetric efficiency which deteriorates combustion process. The HC emission concentration decreases with increasing load because increasing load results in unstable combustion processes due to the increase in combustion temperature associated with higher engine load.

3.1.3 Carbon Monoxide (CO)

The results of CO concentration are shown in **Fig. 6**, it can be seen from this figure that the increase in engine speed improves engine volumetric efficiency and mixing process, leading to better combustion process and this lead to a reduction CO emissions. The CO emission shows the same trend as unburnt hydrocarbon since both are products of incomplete combustion of fuel. At load condition, the concentration of CO was lower as compared with the no-load condition because as turning the air-conditioning system as a load lead to increase in the engine temperature and to more efficient combustion, as a result, less of carbon monoxide production.

3.1.4 Carbon Dioxide (CO₂)

On the other hand, as can be seen in **Fig. 7**, that the increase in engine rotation speed caused a significant increase in CO₂ emission from the exhaust gas, this is due to larger oxidation rate of fuel carbon to CO₂. Air conditioning system led to significant increase in the emission of CO₂ as compared with the normal position, and this behavior was the result of good burn of fuel at high engine temperature. It has been noticed that the concentration of CO₂ after 3000 rpm was increased and it may be due to ICE processes.

3.2 The Effect of NTP Reactor on the Exhaust Gases Emissions

Different values of voltage were applied on to the electrodes of the reactor to generate the plasma. The NTP was created at 10~12kv and that is due to the distance, material, and shape of the electrode.

3.2.1 The Effect of NTP on NO_x Removal

As can be seen in **Fig. 8 (a and b)** the concentration of NO_x decreases slightly before the applied voltage reach 13kv, plasma has not been powerful enough to make significant variation in the NO_x concentration when the voltage less than 13kv. By increasing the applied voltage, the concentration of NO_x decrease due to the occurrence of series reactions in the NTP reactor. It can be noted that the concentration of NO_x at engine rotated speed of 3500rpm was changed



from (112ppm) at an applied voltage of 0kv to (31ppm) at applied voltage of 25kv, so the highest removal efficiency recorded 72.32%.

3.2.2 The Effect of NTP on HC Removal

After 13kv the NTP show a good potential for hydrocarbons reduction. **Fig. 9** shows that the reduction started at a different voltage value for the rotating speeds, for example at 2500 rpm the reduction started at 15kv, while at 4000 rpm started at 17kv as shown in Table 1. The concentration of HC has not been affected too much after an applied voltage of 25kv for all considered rotating speeds. The concentration of HC at engine rotated speed of 1500rpm was changed from (131ppm) at an applied voltage of 0kv to (40ppm) at applied voltage of 30kv, so high removal efficiency was exceed 69.46%.

3.2.3 The Effect of NTP on CO Removal

Fig. 10 (a, b, c and d) shows the CO concentration for different applied voltages and different rotating engine speeds before and after release from the reactor. As displayed, the concentration of CO was very low at the reactor outlet due to low CO content of gases and remained almost little change under plasma treatment. This means that at the given conditions of applied voltage, rotating engine speeds and with respect to the amount of oxygen content of the exhaust, the hydrocarbons were removed and some of them converted to CO₂ more than CO during the experiments. The concentration of CO at an engine speed of 3000rpm was changed from (0.03% vol) at 0kv to (0.01% vol) at the range of (25-32kv) of applied voltages and the removal efficiency was to be 66.66%.

3.2.4 The Effect of NTP on CO₂ Removal

As already discussed, the highest performance of NTP can be achieved at high applied voltage levels. **Fig. 11 (a and b)** shows the variation of the concentration of CO₂ as a function of applied voltage for different rotating engine speeds. The concentration of CO₂ was decreased slightly with the increase of the applied voltage in the range of (0-13kv) for all the studied engine rotating speeds of (750-4000rpm). Moreover, by increasing the voltage above 13kv, the concentration of CO₂ decreases continuously and significantly for the condition of the engine rotating speeds of 3500 and 4000 (rpm). The CO₂ concentration falls from (10.56% vol) at a voltage of 0kv to (2.91% vol) at 27kv of the voltage applied with a removal efficiency of 72.44%.

3.3 Overall Best Performance of NTP in Pollution Reduction

To provide an overall view of the effect of non-thermal plasma on different emission gases, as a final analysis all emission gases have been plotted in **Fig. 12** and **Fig. 13** at different operating conditions. As can be seen in **Fig. 12**, plasma can decrease the concentration of the emissions of NO_x and HC. It can be observed that the concentration of NO_x at engine rotated speed of 3500rpm is changed from (112ppm) at an applied voltage of 0kv to (31ppm) at an applied voltage of 25kv, so the removal efficiency recorded 72.32%. The concentration of HC at engine rotating speed of 1500rpm is changed from (131ppm) at an applied voltage of 0kv to (40ppm) at an applied voltage of 30kv, so the removal efficiency exceeds 69.46%.

Fig. 13 shows the concentration of CO at an engine speed of 3000rpm is changed from (0.03% vol) at 0kv to (0.01% vol) at the range of (25-32kv) of applied voltages and the removal efficiency been 66.66%. At last the CO₂ concentration has fallen from (10.56% vol) at a voltage of 0kv to (2.91% vol) at 27kv of the voltage applied with a removal efficiency of 72.44%.



4. CONCLUSIONS

1-In the normal mode of the engine exhaust, it has been found that the concentration of NO_x and CO₂ increase as the engine rotation speed increase for the two engine conditions.

2-HC and CO concentration is decreased as the engine rotation speed increase in case of no-load and with load condition.

3-NO_x removal efficiency is increased when increasing the applied voltage to the NTP reactor, to exceed 72.32% removal at 3500 rpm with a voltage of 25kv.

4-NTP is found also very effective for HC removal, 69.46% removal at 1500 rpm and an applied voltage of 30kv.

5-Generally, the best decomposition processes for CO and CO₂ is observed at low and high rotation engine speeds, the higher removal for CO₂ concentration is 72.44% at 3500 rpm and 27kV.

6-CO concentration is to be from 0 % to 0.07 %, so a small CO decomposition was observed in plasma treatment and that due to the low initial concentration rate resulted from the engine exhaust, best removal rate was to be 66.66% at 3000 rpm and a range of voltage (25-32kv).

5. ACKNOWLEDGMENT

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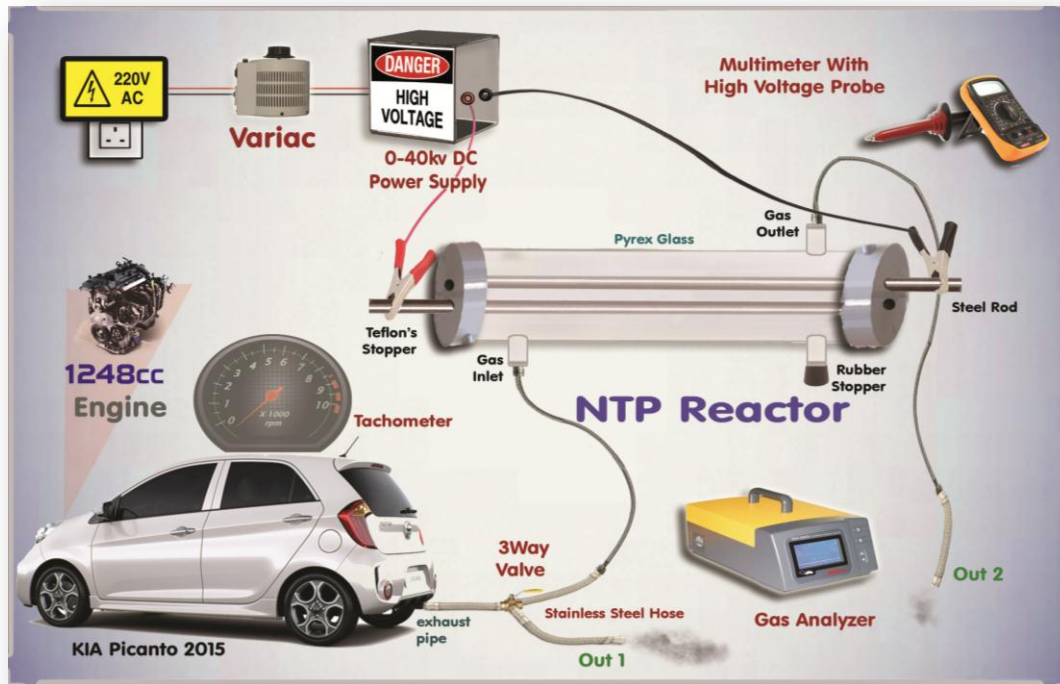
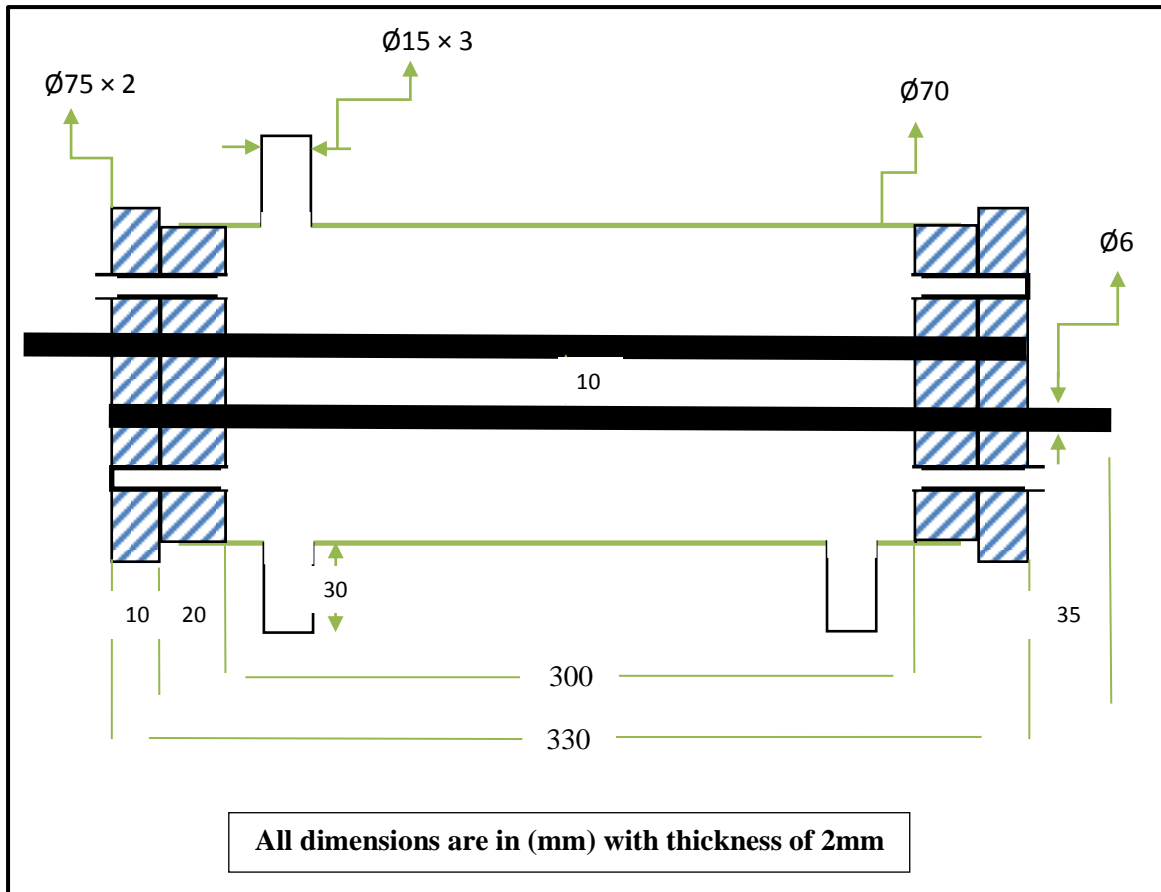


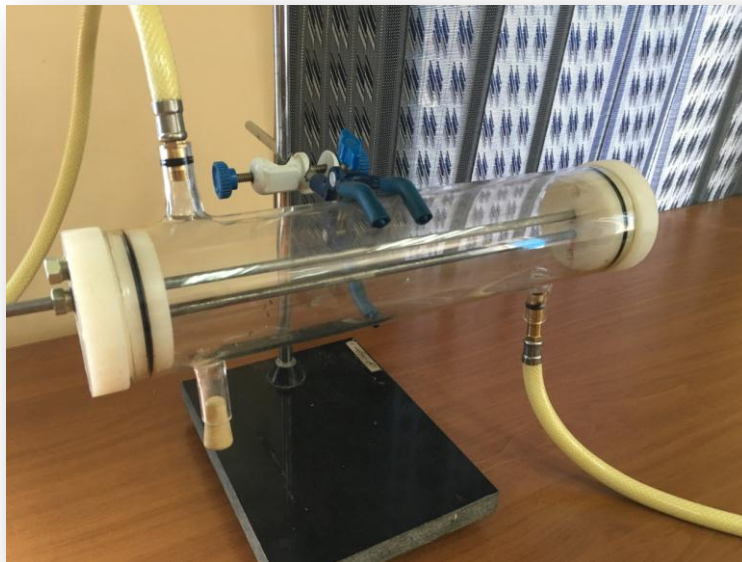
Figure 1. Schematic diagram of the experimental apparatus.



Figure 2. Photograph of experimental apparatus.



(a)



(b)

Figure 3. (a) Cross-sectional view of designed Nonthermal plasma.

(b) Photograph of Non-Thermal Plasma Reactor.

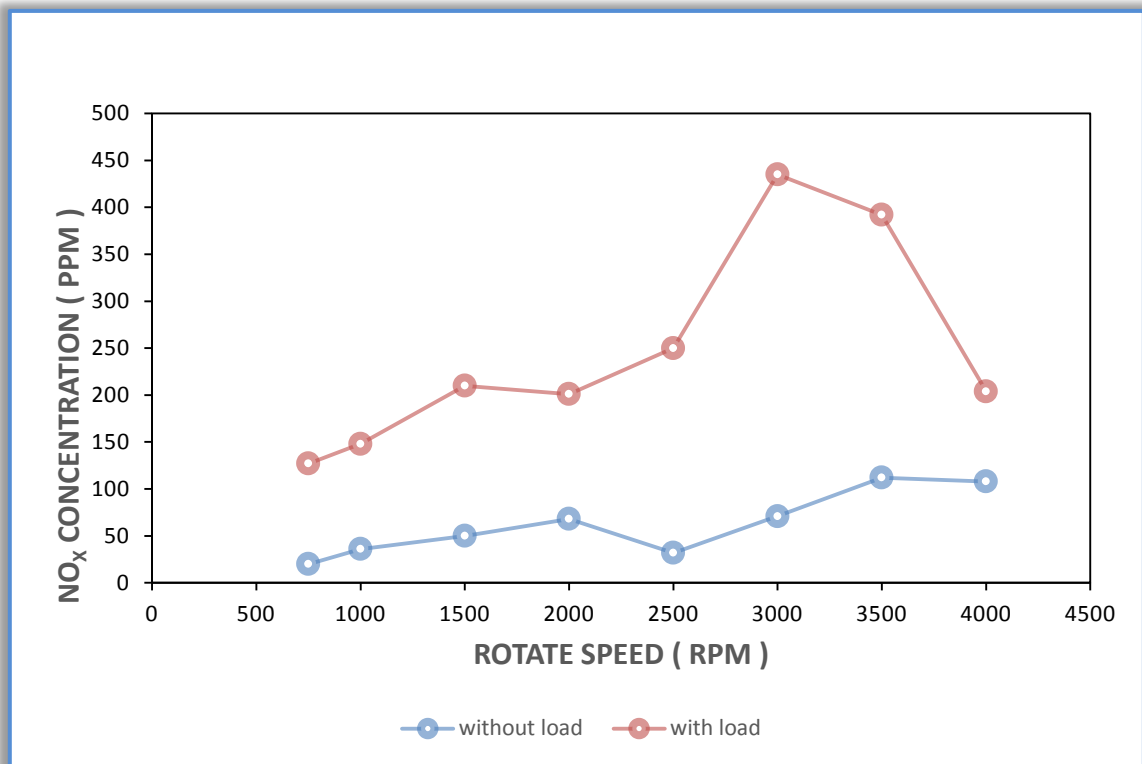


Figure 4. NO_x concentration against engine rotated speed at the motor condition.

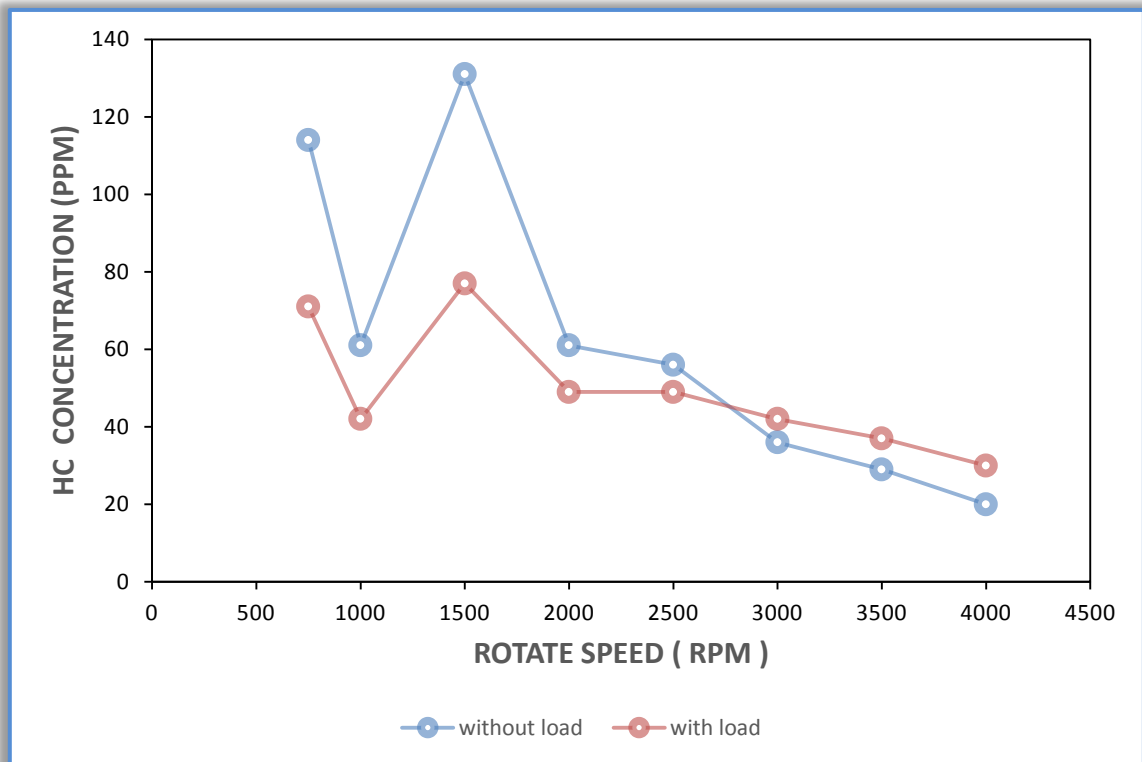


Figure 5. HC concentration against engine rotated speed at the motor condition.

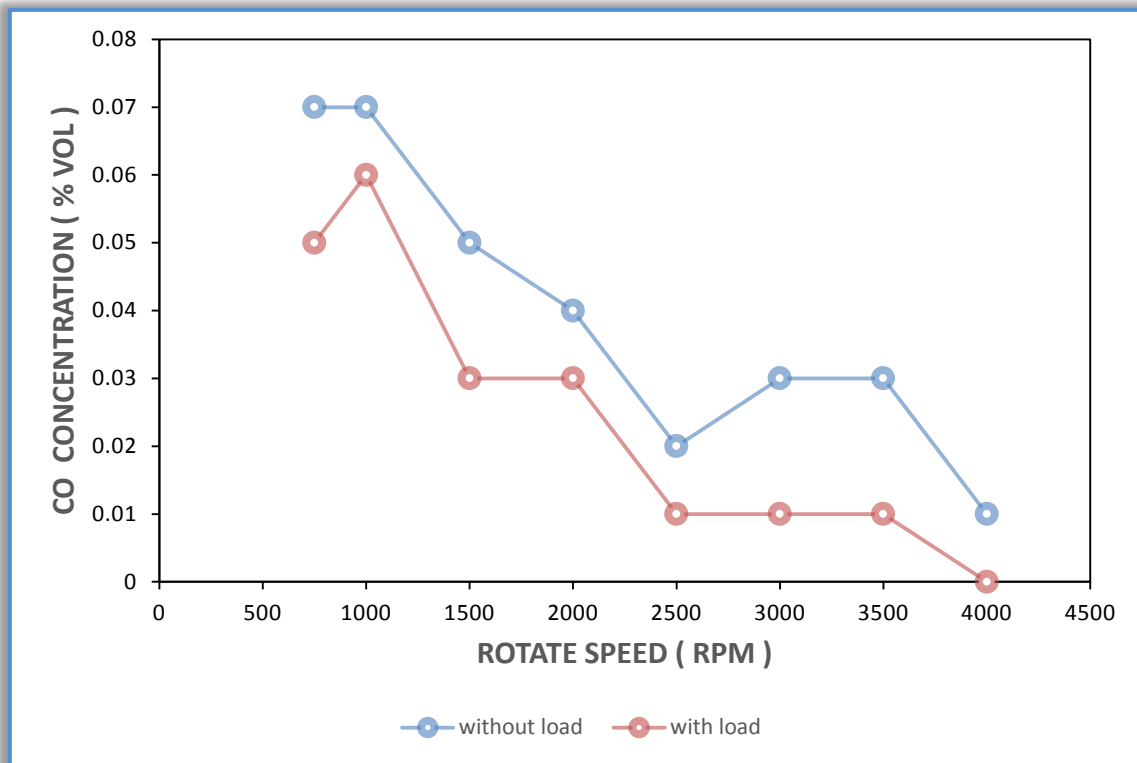


Figure 6. CO concentration against engine rotated speed at the motor condition.

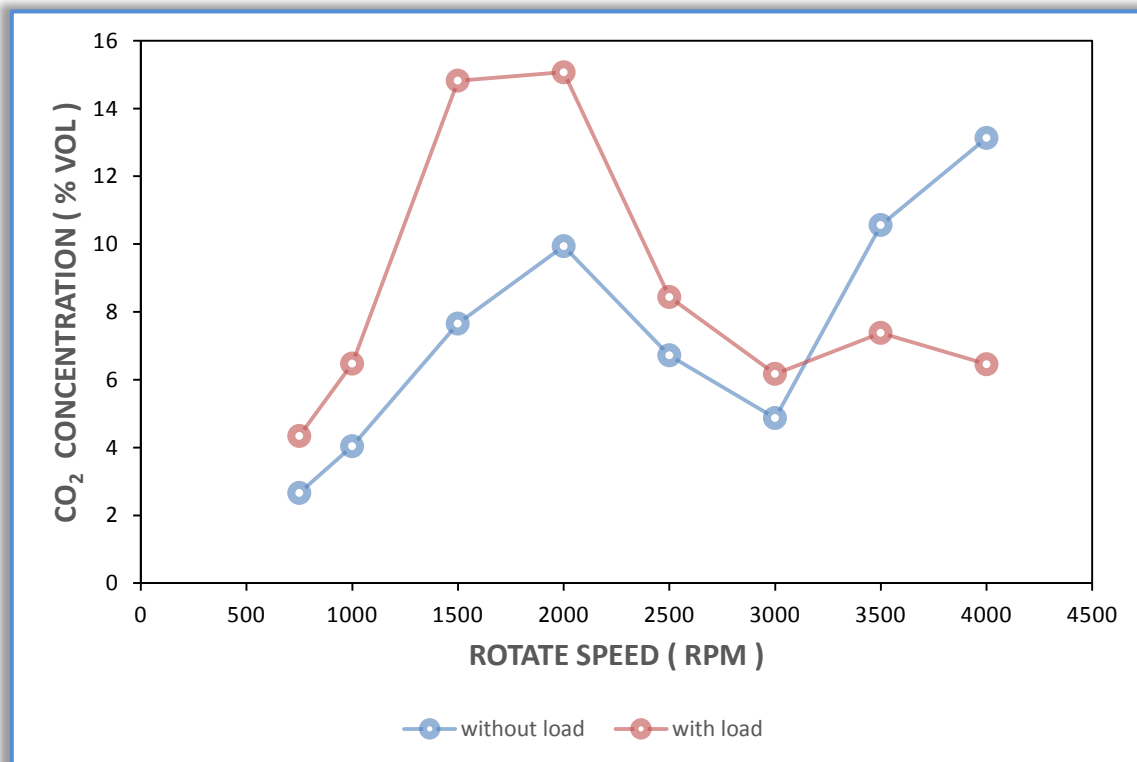
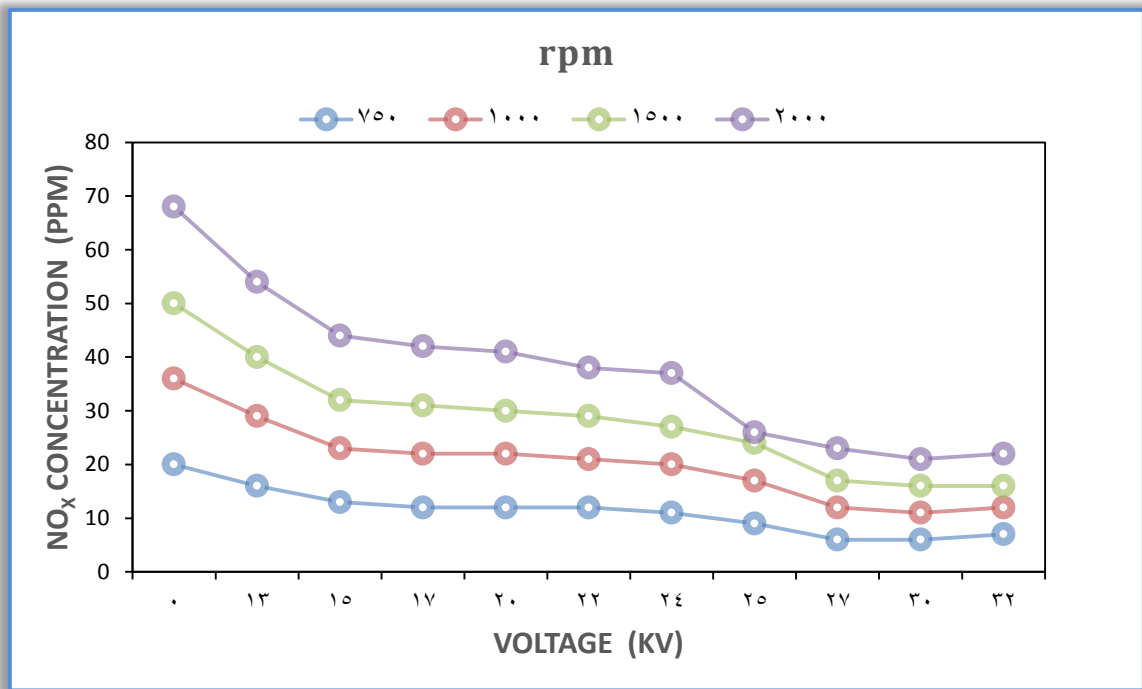
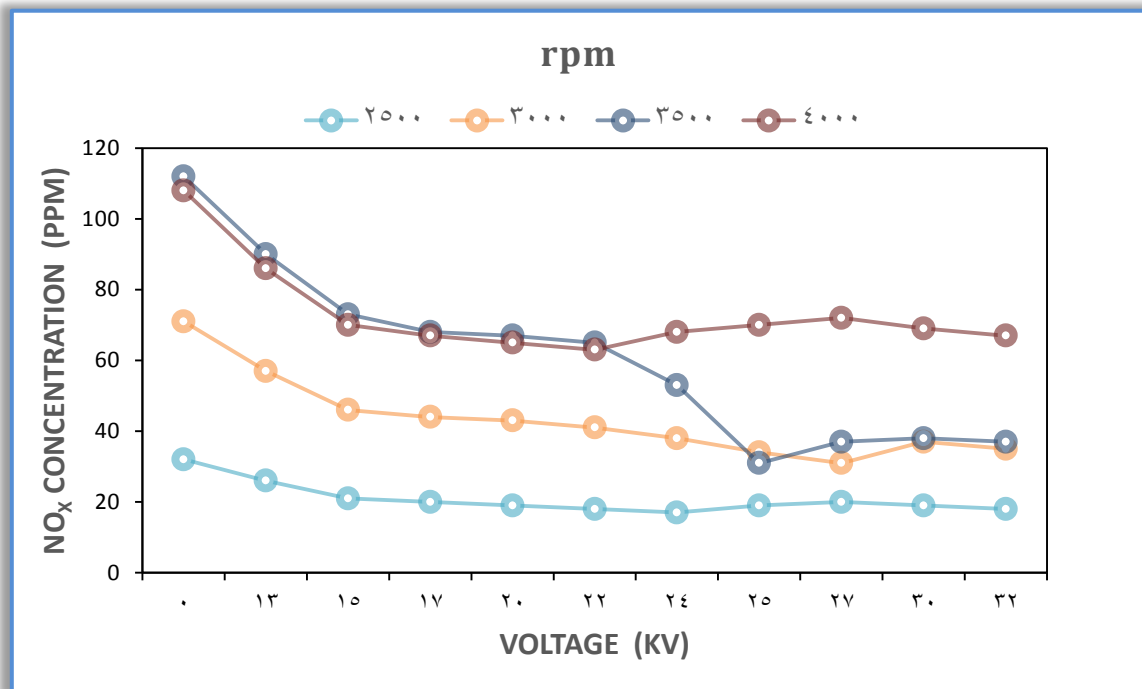


Figure 7. CO₂ concentration against engine rotated speed at the motor condition.



(a)



(b)

Figure 8. (a and b) NO_x degradation at different voltage values.

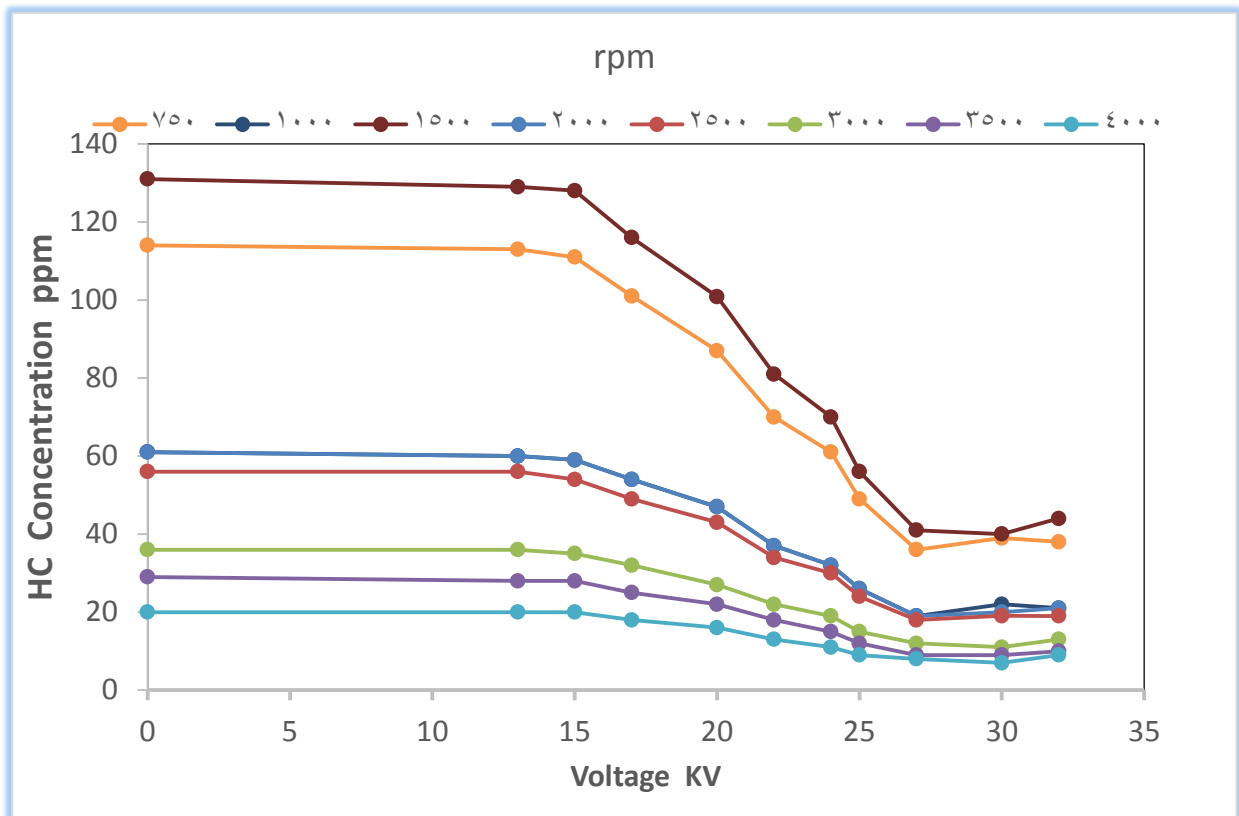
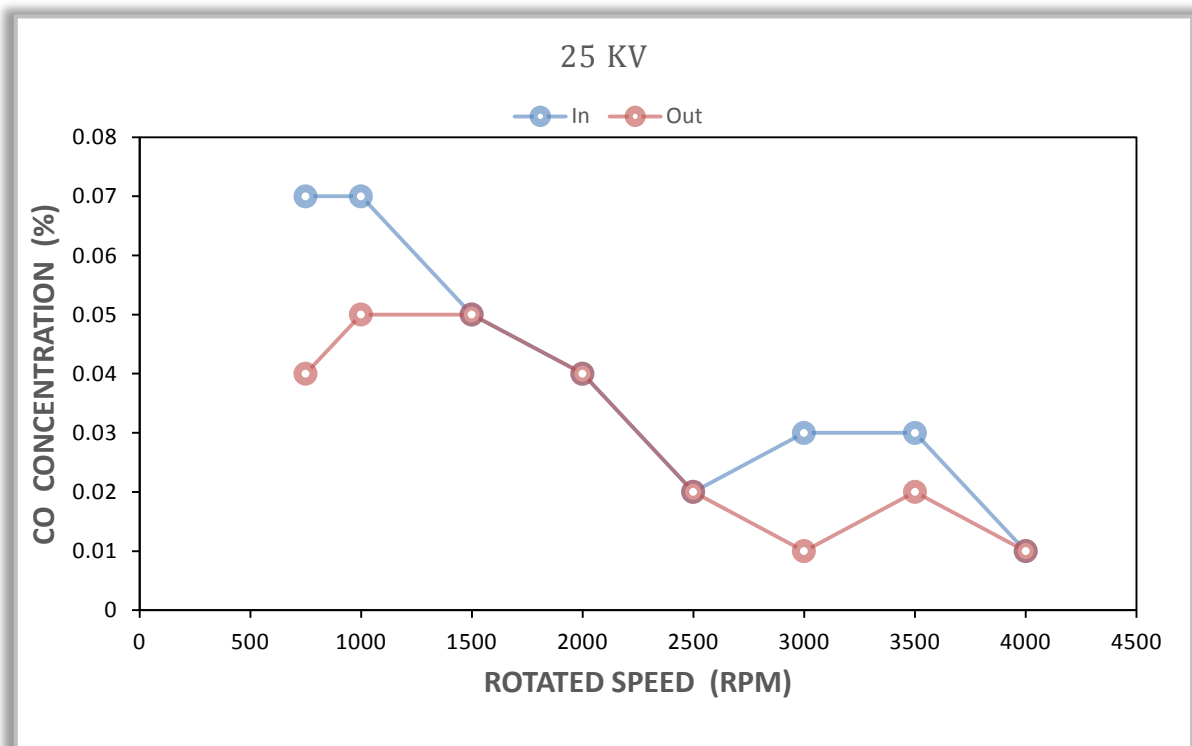
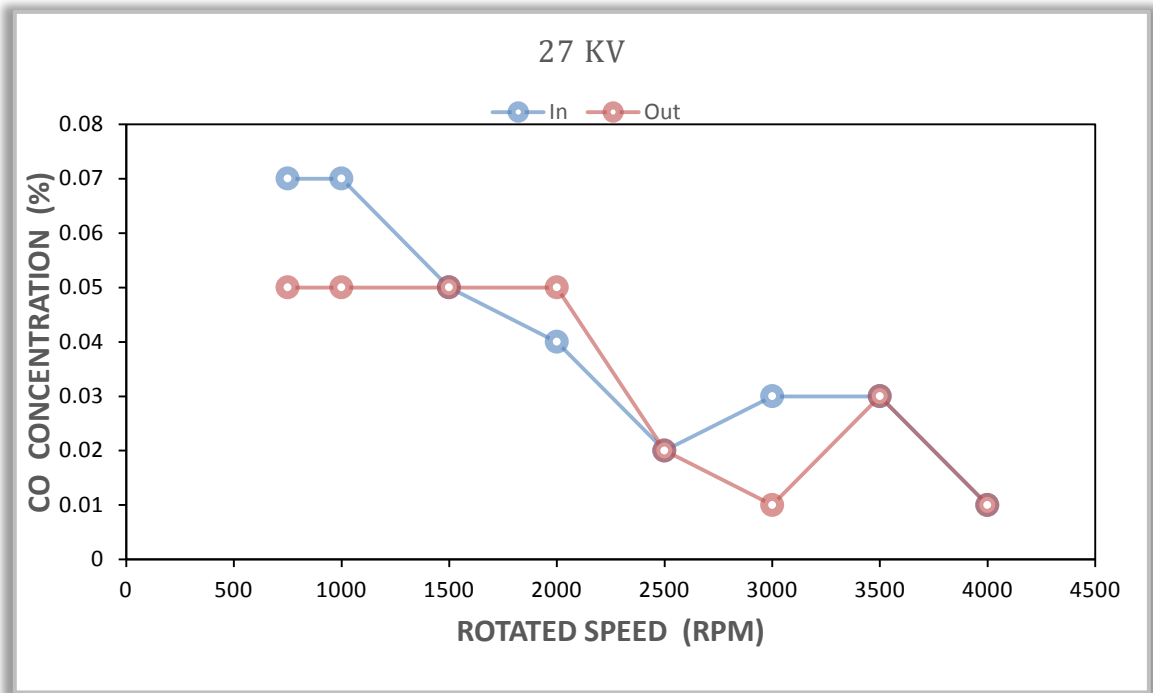


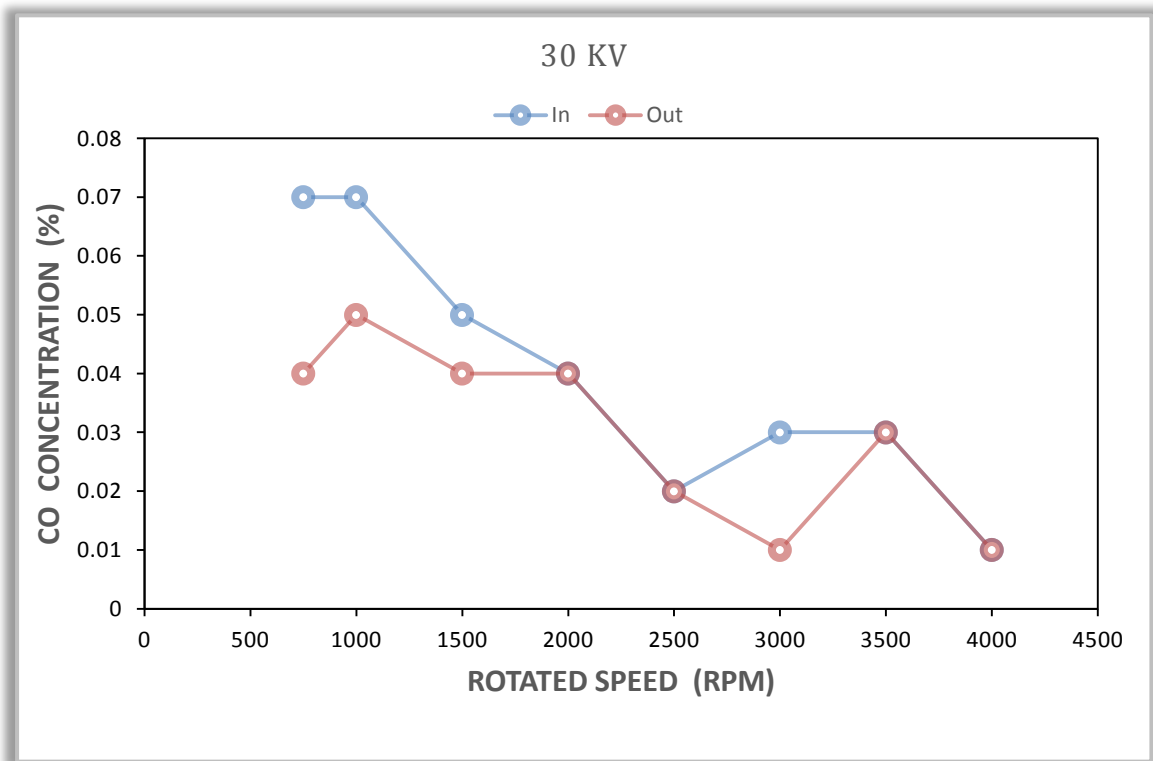
Figure 9. HC degradation at different voltage values.



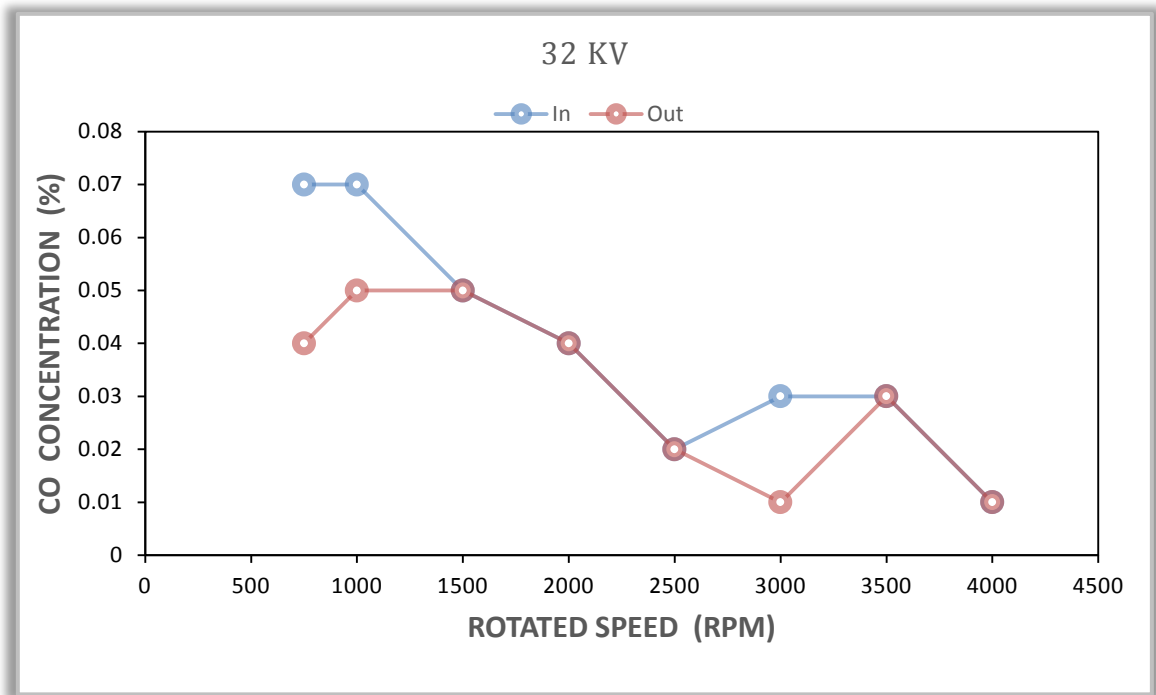
(a)



(b)

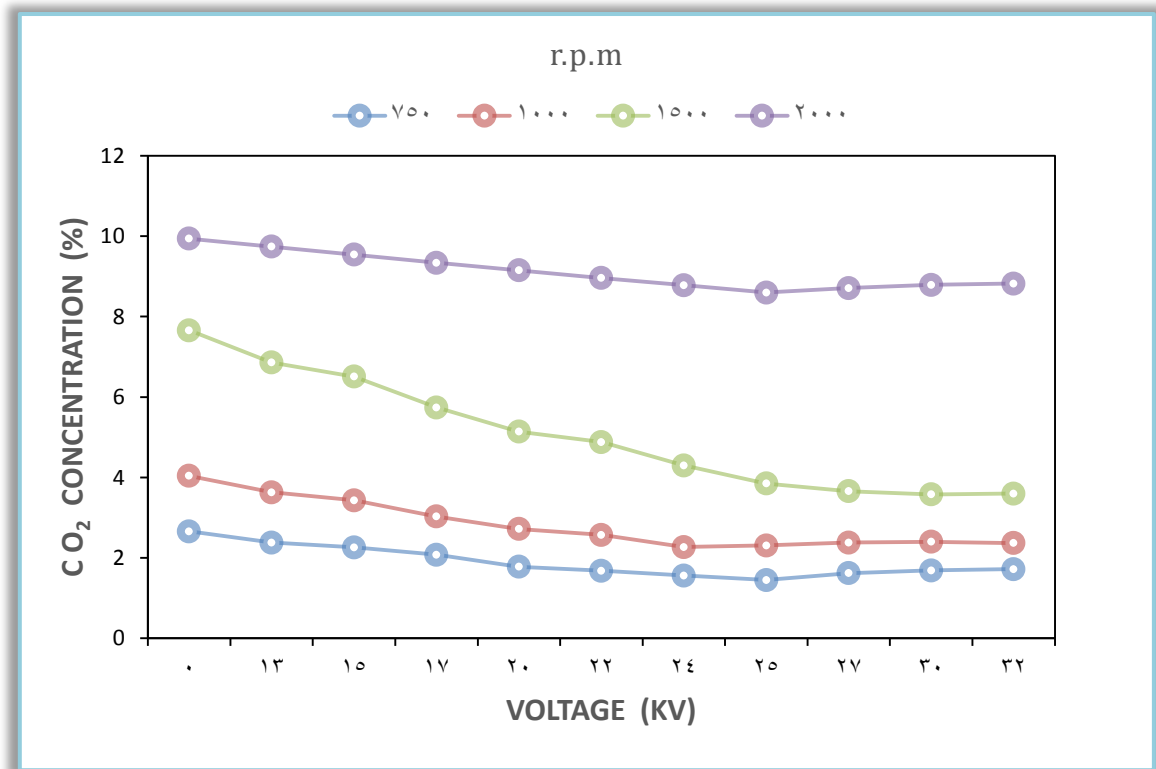


(c)

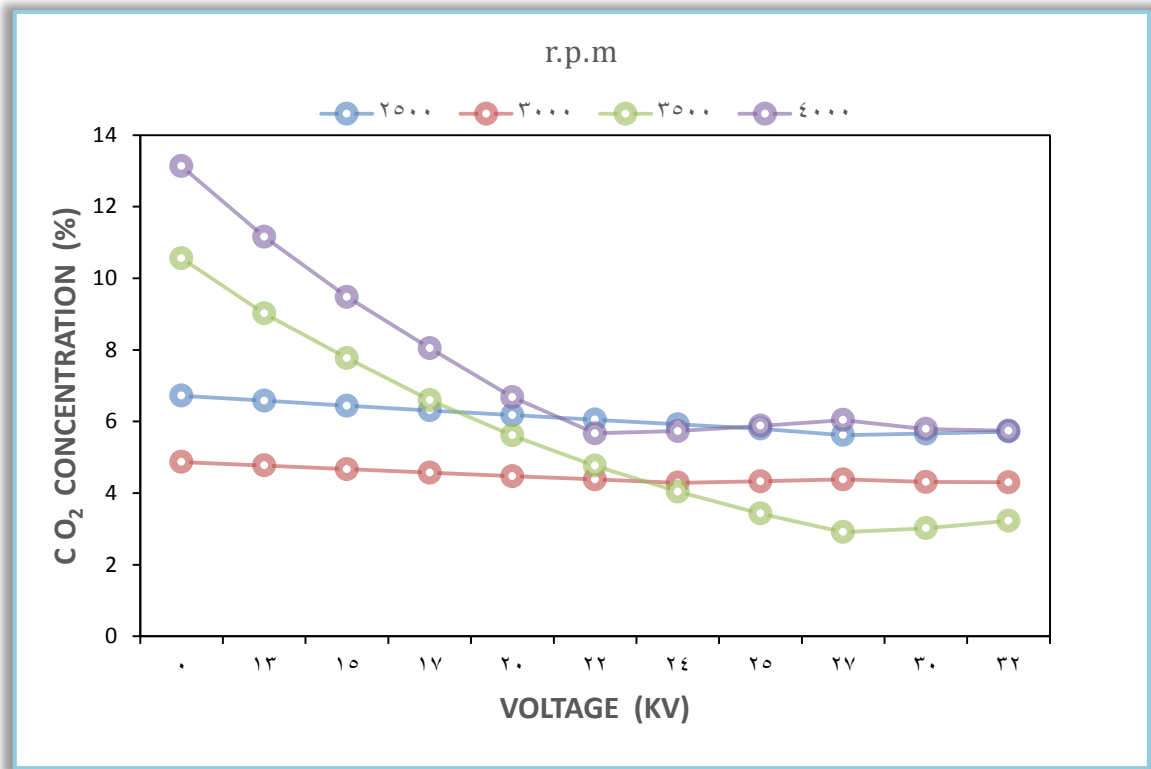


(d)

Figure 10. (a, b, c and d) CO degradation at different voltage values.



(a)



(b)

Figure 11. (a and b) CO₂ degradation at different voltage

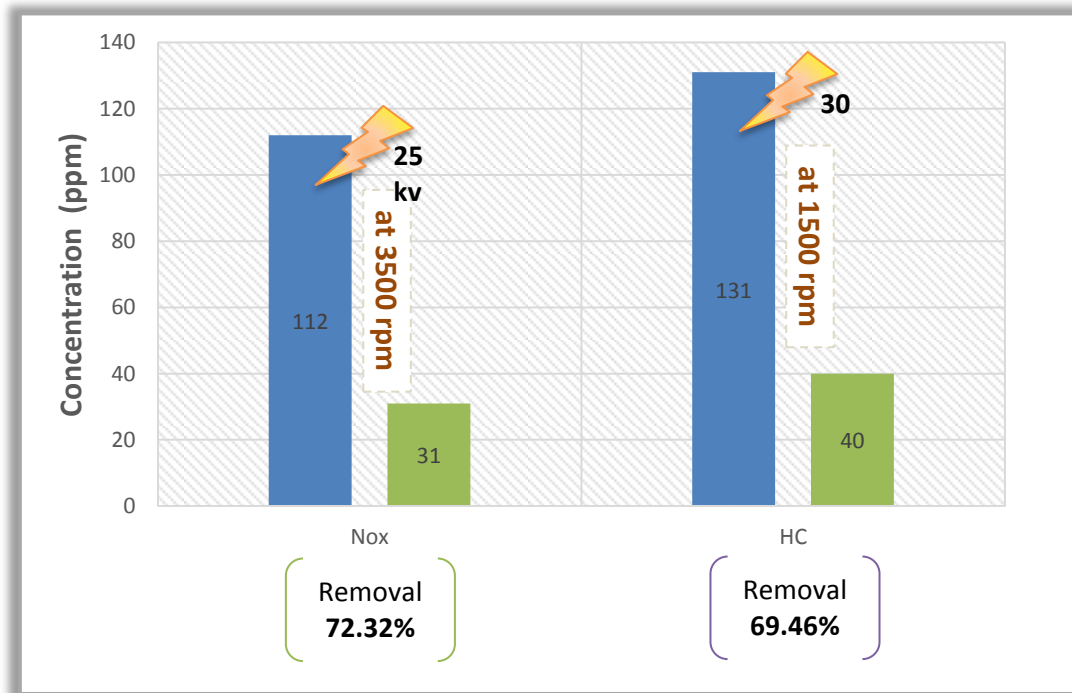


Figure 12. Best performance of NTP on NO_x and HC.

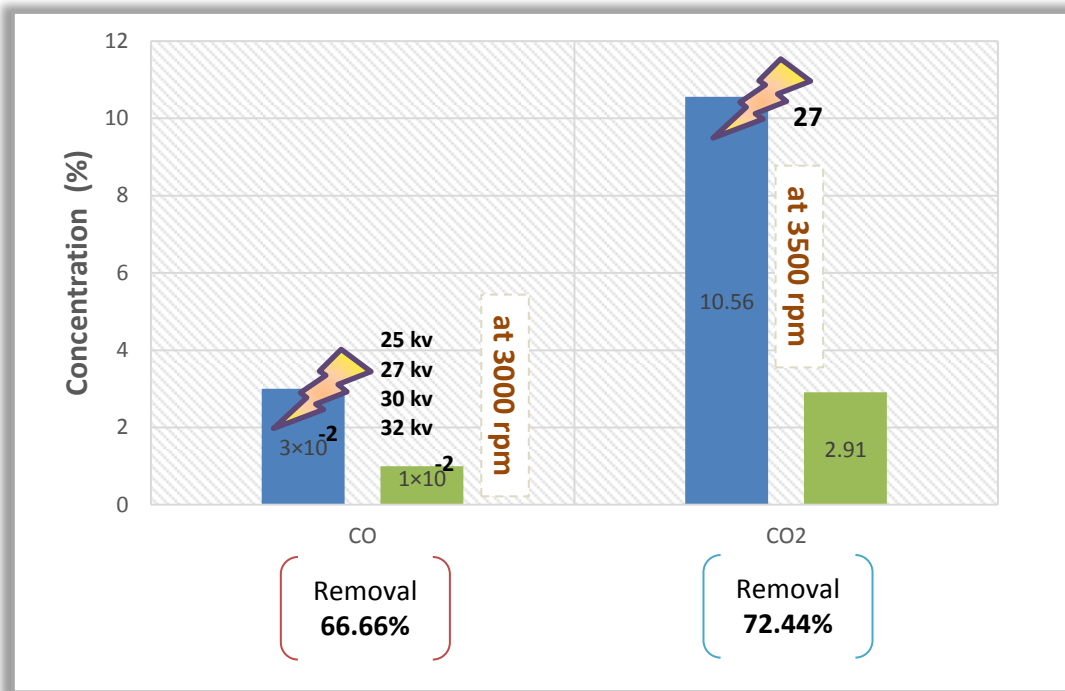


Figure 13. Best performance of NTP on CO and CO₂.

Table 1. HC concentration readings at different rotated speed and voltage values.

rpm	hc(ppm)	13kv	15kv	17kv	20kv	22kv	24kv	25kv	27kv	30kv	32kv
750	114	113	111	101	87	70	61	49	36	39	38
1000	61	60	59	54	47	37	32	26	19	22	21
1500	131	129	128	116	101	81	70	56	41	40	44
2000	61	60	59	54	47	37	32	26	19	20	21
2500	56	56	54	49	43	34	30	24	18	19	19
3000	36	36	35	32	27	22	19	15	12	11	13
3500	29	28	28	25	22	18	15	12	9	9	10
4000	20	20	20	18	16	13	11	9	8	7	9