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ORIGINAL ARTICLE

Development of Air Treatment Technology Using Plasma Method

RASOUL YARAHMADI^{1,} SAYED BAGER MORTAZAVI^{2*}, and PARVIN MORIDI³

¹Department of Occupational Health, Research Center for Occupational Health, School of Public Health, Tehran University of Medical Sciences, Tehran, Iran; ²Department of Occupational Health, Faculty of Medical Science, Tarbiat Modares University, Tehran, Iran; ³Department of Environment Management, Azad University of Researches and Sciences Branch, Tehran, Iran

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ABSTRACT

Due to the physicochemical properties of nitrogen oxides as active molecules, the removal of this group of pollutants has always been considered as a matter of concern for specialists. The present study seeks to develop the removal technique of nitrogen oxides "as a major type of air pollutant" by means of nonthermal plasma process under atmospheric conditions. Besides having the potential to reduce energy consumption in the pollutant removal process, non-thermal plasma technology also provides particular flexibility for the simultaneous removal and mitigation of the secondary pollutants. In this research we have used the Dielectric Barrier Discharge (DBD) plasma process to achieve an effective conversion of Nitrogen Oxides (NOx). As a result of the collisions between the electrons and the airflow containing NOx, active radicals and molecules are generated in a limited and controlled volume (plasma reactor) and the conversion and removal process is then carried out in the presence of hydrocarbon as reducer gas. The key factors for NOx conversion especially in the non-thermal plasma condition are the geometric structure and design of the reactor, type of discharge, type of power supply, temperature, space velocity, propane/NOx mole ratio, and voltage. In the present study, the factors of temperature, mole ratio of the reducer and input voltage were examined. The results showed that the optimal conditions for conversion of NOx into N₂ and O₂ are temperature of 180°C, propane/NOx mole ratio of 0.5, and voltage of 5 KV. Under the optimal conditions acquired, NOx conversion was 0.79 in the 100 PPM concentration. Under the conditions of the present study, 1-5 PPM Ozone, Formaldehyde, and CO was generated as the undesirable pollutants.

Keywords: Nitrogen Oxides, Non-Thermal Plasma, Dielectric, Air Treatment, Optimal Conversion

INTRODUCTION

A partially or completely ionized gas containing free electrons, ions, and radicals is called plasma. The technical application of plasma is expanding increasingly and this is due to its special characteristics and technological nature. The most important influential factors in plasma performance are the parameters and variables of the plasma system [1-3].

The high emission of gaseous contaminants into the living and working environment has caused irreparable effects on humans, plants, soil and water. Furthermore, adhering to the national and international obligations and commitments in the form of protocols and approvals issued on the mitigation of the emitted pollutants has multiplied the importance of controlling the NOx as the second atmospheric pollutant after the

^{*} Corresponding author: Sayed Bager Mortazavi, E-mail: mortazav@modares.ac.ir

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Fig 1. Plasma reactor schematic. a: Thermal sensor b: Holder between cathode and anode c: Quartze dielectric barrier discharge d: tangestan wire as cathode e: cuoper shield as anode and reactor main body f: Anode connected to earth



Fig 2. Set up of experimental design. Mass flow controller (MFC), primery mixing zone (PMZ), secondary mixing zone (SMZ)

airborne particles [4]. Among the NOx contamination controlling techniques including selective, nonselective, and chemical reduction, and plasma-aided treatment, each has its own features and potentialities and is of great importance due to the simplicity of the treatment process, the low cost of designing, manufacturing and maintenance, no need for mechanical and chemical equipments, and also innovation of plasma technology as a preferable method for air treatment in the latest decade [3, 5].

The control of velocity and space time of the exhaust gas from the fixed and removable sources is so significant and effective in mixing and completing the final reactions of the plasma treatment that we can use this potentiality with minimum cost and considering the technical and economical aspects. Due to the technical considerations in designing the geometric structure of plasma reactor, the two key factors of pressure drop and dead volume, as effective factors in reducing the treatment output, will modification [6-8].

The main aim of this study was the identification, modification, and optimization of the physical and chemical parameters and variables for the optimal removal of the nitrogen oxide contamination in exhaust gases. Therefore, to improve the technical knowledge of these kinds of technologies, a better comprehension of key and basic considerations is required [9].

MATERIALS AND METHODS

A bench scale apparatus composed of non-thermal plasma reactor, thermal furnace, power supplies of selected gases, gas measuring stations, NOx and water vapor (Figs 1 & 2) was utilized.

Temperature setting

The input temperature of the plasma reactor is first measured by thermal sensors and then transmitted to the furnace thermal monitoring system (thermocouple TC_1 10°C per min, with an accuracy of 0.1°C) to be controlled. In this study, a dielectric barrier discharge system is used in non-thermal plasma process under atmospheric conditions in 100-180°C range.

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Temperature	Voltage	3 KV				KV5				KV7				10 KV			
	NO _X C ₃ H ₈ / NOx CONVERSION	0.3	0.5	1	1.5	0.3	0.5	1	1.5	0.3	0.5	1	1.5	0.3	0.5	1	1.5
100° C	NO	11.6	.221	90	93	51.4	51.2	5.69	76	33	32	56	17	81	63	22	94
	NOx	28	21	57	34	32	21	38	35	20	20	10	17	35	9	17	51
140° C	NO	39	35	29	51	80	16	30	83	56	28	33	84	43	50	61	miss
	NOx	37	40	30	58	50	9	23	47	39	33	20	49	22	42	55	75
180° C	NO	57	66	37	41	82	83	67	47/9	-5	98	67	52	-10	89	74	80
	NOx	52	60	48	38	35	79	51	43	8/4	46	50	46	48	68	53	64
220° C	NO	43	48	43	47	54	52	53	70	53/8	61	53	68	52/7	64	53	64
	NOx	39	56	35	40	40	37	53	47	41	42	43	44	39	42	-6	36

Table 1. Effect of propane/NOx mole ratio and temperature interaction on NOx Conversion in to N_2 with various reducer gas concentration and input voltage in NTP

Note 1: Under the conditions of study, 1-5 PPM ozone, formaldehyde, and CO was generated as the by-products pollutants.

Note 2: NOx gas contain 10% NO₂ and 90% NO

Feed gas stream

The feeding system is supplied by oxygen and nitrogen capsules with the percentage purity of 99.99% and microcapsules containing nitrogen dioxide and monoxide and propane (micro gas) at a certain volumetric concentration. The system input air volume is regulated by the Mass Flow Controller (MFC) and then conducted to mixture and thermal units. The connectors are selected by appropriate materials resistant to heat and pressure with attention to the considerations on temperature and pressure of the injected flow.

Power supply

The plasma power supply is an AC generator with a voltage of up to 20 KV, maximum current and detection limit are 20 and 0.01 mili ampler.

Experimental Design

In order to achieve more accurate results with a higher level of significance, the experiments were carried out on the three major factors of temperature, voltage and mole ratio of propane, first at two levels¹ in order to range finding, and then at four levels $(4^k)^2$ [10].

Sampling points

The aim of this method is to obtain accurate and reasonable information and results through a practical experimental design. Products from the decomposition and interaction of the system inlet gases were measured at sample points before and after the reactor via two methods and with two objectives. In the first method, with the quantitative evaluation objectives, an instant analysis of the sampling points was performed by a direct-reading gas detection instrument³. In the second method, with quantitative and qualitative objectives, after collecting grap samples in the glass containers installed in parallel along with direct-reading sampler, an analysis of all possible parameters and annoying and interfere or undesirable factors was carried out by Gas Chromatography with Spectrometric GC-MS. Results of the second objective and the comprehensive analytical method would lead us to use the mass balance of the contaminants under study.

To establish a mass balance before and after the plasma reactor, all volumetric concentrations (PPM_v) in the system flow rate were converted into the mass flow (mg/min) and NO and NOx conversion efficiency was then calculated and evaluated separately.

Previous studies indicated that the amount of humidity and oxygen in non-thermal plasma process usually affects removal and conversion of NOx contamination in a constant limit; on this basis, an amount of 5% oxygen and 10% humidity is considered as invariable in all experiments [11, 12].

In this research, an airflow with 100 PPM_v NOx volumetric concentration (90% NO, 10% (NO₂) and 2.5-5 LPM flow rate was used.

The data analysis was performed by SPSS and Minitab 14 software. The research hypotheses were tested through *t*-test, Correlation, Paired *t*-test, two-way-ANOVA, and the minitab 2D graphs.

RESULTS

In the present research, a multi-way ANOVA general⁴ with a confidence level of more than 95% and significance level of less than 0.05 was performed on

¹ Two-level factorial design

²Four-level factorial design

³ MRU Air, Industrial Varioplus

⁴ Linear model univariate(GLM)



Fig 3. Effect of voltage and reducer gas interaction in non-thermal plasma on NO Conversion at 100 degree selsius



Fig 4. Effect of voltage and reducer gas interaction in non-thermal plasma on NO Conversion at 140 degree selsius

conversion of oxides of nitrogen (NO, NOx) as the response variable.

A four-level factorial experiment was carried on response variable (NOx Conversion), temperature in the range of 100-180°C, Voltage in the range of 5-10KV, propane/NOx mole ratio in the range of 0.3-1.5.

Table 1 shows the results of the interaction of effective factors on NO_x contamination conversion in plasma reactor. As indicated in the table, the interaction of temperature, voltage difference, and reducer gas affect the NOx, NO conversion. The voltage factor has relatively increased NOx conversion levels in the region of 5KV (the initial production level of corona). The fourth level of temperature (220°C) undergoes a

minimal effect, as "NOx Conversion", due to the rapid oxidation of propane.

In order to locate the region of the optimum for NOx conversion under the conditions of the present experiment, a comparison of equality of variances of the response variable (NOx conversion) was performed at four levels of temperature. The test results with the confidence level of 95% confirm that there is no difference between the variances at the four temperature levels. Furthermore, plasma threshold begins at 5KV [13] hence, because of the relatively high NOx conversion ratio at 180°C which is near the output temperature of mobile source emissions (automotive) the intersection of the two factors can locate the



Fig 5. Effect of voltage and reducer gas interaction in non-thermal plasma on NO Conversion at 180 degree selsius



Fig 6. Effect of voltage and reducer gas interaction in non-thermal plasma on NO Conversion at 220 degree selsius

approximate region of the optimum for NOx conversion.in this study however, the more exact region and points is located by examining the plasma reactor process considerations (minimum energy consumption, minimum reducer and maximum contamination conversion). Figs. 3-10 show the results of the interaction of the factors under study on the response variable (NOx conversion).

DISCUSSIONS

The interaction behavior of the three factors under study on NOx conversion in plasma reactor was examined in this research. Results of a four-level factorial experiment (4^k) with a confidence level of more that %95confirmed that the four levels of temperature have the same effect on NO conversion (Table 1).

The comparison of the process of NOx pollution conversion in non thermal plasma reactor (100°C &140°C): As noted above the interaction of the three factors of temperature, propane and voltage was examined on the airflow with a mass flow of 5 LPM and NOx concentration of 100 PPM (NO 90%, NO₂ 10%); each factor was examined at four levels $(4^k)^1$ under nonthermal plasma condition with NO and NO_x being examined separately (Figs. 3-10). An increase in the propane mole ratio and thus the reducer gas density under non-thermal plasma condition decreases the NO_x concentration in the reactor outlet. The results confirm the positive effect of propane on conversion and removal of NOx contamination. The researches indicate

¹Four Levels Factorial



Fig 7. Effect of voltage and reducer gas interaction in non-thermal plasma on NOx Conversion at 100 degree selsius



Fig 8. Effect of voltage and reducer gas interaction in non-thermal plasma on NOx Conversion at 140 degree selsius

that the generation of high energetic electrons in a plasma reactor does not depend on the field intensity and voltage consumption; rather, the significant parameter for electron's energy in the discharge field depends on the initial voltage (corona threshold). Results of the studies also indicate that the low rate of oxidation NO+HC_s+O₂ \rightarrow NO₂+CO₂+H₂O leads to the generation of by-products such as HCHO, CO and NO₂, while the rapid oxidation and reduction in plasma processes result in the formation of stable molecules and stabilization of radical reaction [14].

At a voltage of 3 KV (before corona threshold), the interaction between system input molecules (oxidation and reduction) occurs under temperature condition, in space time, and in presence of oxygen and water vapor. The relation below shows the nitrogen oxide conversion under non-plasma condition.

Gas phase oxidation:
$$O+NO+M \rightarrow NO_2+M$$
 (2)
 $NO_2+O \rightarrow NO+O_2$ (3)

Likewise, relation below shows the nitrogen oxide conversion under non-thermal plasma

$$e+O_2 \rightarrow e+2O \tag{4}$$

$$N_2 + e \rightarrow N + N + e$$
(5)
$$NO + N \rightarrow N_2 + e$$
(6)

Also the NO_2 will be generated through the reverse reaction as follows

$$2NO+O_2 \rightarrow 2NO_2 \tag{7}$$

$$NO+O_3 \rightarrow NO_2+O_2 \tag{8}$$

Reaction strongly depend on the gas temperature [15]. The slow rate of NO & NOx conversion under the equal conditions of 3 KV and 100°C indicates that NOx conversion is greater than NO (Figs. 3 & 7). Results of the studies on NOx removal confirm the conversion of NO2 into N2 under non plasma condition with no need for radical reactions [16, 17].

NO (g)
$$\rightarrow \frac{1}{2}N_2 + \frac{1}{2}O_2$$
 -20.7 KCal/ mol at 25 C (1)



Fig 9. Effect of voltage and reducer gas interaction in non-thermal plasma on NOx Conversion at 180 degree selsius



Fig 10. Effect of voltage and reducer gas interaction in non-thermal plasma on NOx Conversion at 220 degree selsius

Likewise, at a voltage of 5 KV, because of entering the phase of the minimum conditions for the excitation and decomposition of input matrix molecules, and NO high oxidation potential under corona condition, we see that by an increase in the mole ratio of the reducer, NO conversion ratio varies between 51.2 and 86 percent; while NOx conversion is 38% maximum. On this basis, to confirm the hypothesis of the high efficiency of nonthermal plasma in optimal condition, results of NO into NO₂ conversion provided an appropriate response. The slow rate of NO_x conversion in plasma (5KV & 100°C) is due to the inhibition of hydroxyl and hydrocarbon free radicals by the $-NO_2$ and R-NO₂ stray ions, while with regard to NO conversion, the products are NO₂ molecules and radicals. At a temperature of 100°C and voltage of 7 KV, due to a higher energy density of molecules and consequently the excitation of the reducer, and water and oxygen molecules, the conversion is obtained with a higher velocity in the reactor electric field. Accordingly, the possibility of the generation of undesirable sustainable products (OH₂, C₂H₄, HCHO, NHO₃, and NO₃) and the nonparticipation in NOx oxidation and reduction reactions causes a drop in conversion efficiency of NOx active molecules.

But at the temperature of 100°C and voltage of 10 KV, doubling the discharge energy in plasma process increases the formation of active radicals such as O_2 , N_2 , CO, and CO_2 as compared with low voltages. It should be noted that although in this case, too, a relatively high NO (94 percent) and NOx (51%) conversion is

obtained, energy consumption per volume of the reactor inflow is doubled.

Figures 4 & 8 illustrate the comparison between the results of NO and NOx conversion and removal at the temperature of 140°C and with different propane mole ratios. The temperature of 140°C, too, provides appropriate conditions for chemical reactions and effective interactions in nitrogen oxides conversion and removal processes. Based on the results, it is possible to deduce that a propane mole ratio of 1.5 ensures a better conversion condition as compared with the other three of the reducer because of levels providing stoichiometric conditions and higher density matrix. Other levels of propane gas, due to the energy levels of active radicals and ions generated during the process and the inadequacy of heat, failed completing final reduction reactions of propane potential convergence in treatment process (<140°C) to the extent that at a voltage of 7 KV, the emission of ultraviolet radiation at quantum level and the electrical discharge energy intensified the voltage in the region of 7 KV (similar to plasma initial threshold). The convergence of the results obtained from this section and those from the study on NOx removal has been cited in some scientific researches [15]. At the voltage of 10 KV and the higher, the concentration of NO gas, as undesirable byproducts, increases due to the conversion of N2O present in the atmosphere and plasma environment into NO (37-76%) [18-20].

The comparison of NOx conversion process in plasma reactor (180°C) The comparison between NO₂ and NO conversion processes affected by the factors of voltage, propane molar ratio at the temperature of 180° C is presented in figure 5 and 9.

At 180°C, NOx conversion is achieved under a natural and non-plasma conditions prior to the plasma production, i.e., at 3 KV, because the conditions required for radical production, oxidation and reduction reactions are not met; so It can be said that the space time and reactor discharge volume has provided the opportunity for a maximum interaction of NOx molecules with a minimum undesirable productions. At corona initial threshold voltage likewise, a propane ratio of 0.3 and 0.5, equal to 50 PPM and 30 PPM, leads to an optimal conversion equal to 82-83 percent within the space time of 0.01- 0.147 second. In the non-thermal reactor plasma (NTP) process, the burn rate (of the reducer gas) and the production rate of active and excited components (ions- radicals) is initiated by a minimum amount of interstitial material. In DBD plasma, the mild collision between the electrons and the reducer, i.e., hydrocarbons, H2O, O2 results in the production of free radicals and ions during a short time interval (2-3 nanoseconds) under optimal energy conditions (1-10 ev). In such circumstances, the heat required for the formation of active compounds reduces as compared with non-plasma conditions [21, 22].

Under the same conditions (input matrix components and temperature of 180°C), the process of NO conversion is accompanied with an increase of voltage from 5 to 7KV, due to a decrease in radical

production (increase in energy density), and a decrease in NO into NO_2 conversion. Results of some scientific researches confirm what was mentioned in this section [23].

The same procedure also occurs in the case of NOx (Fig 9). In this phase of the experiments likewise, the maximum and minimum conversion occurred at corona initial threshold (79%) and at the voltage of 7KV respectively. In both NO and NO₂ cases, by increasing the voltage (twice the corona threshold) nearly all matrixes are converted to active radicals and ions due to the high energy density of electrons. But, as already noted, voltages above the corona threshold accelerates the N₂O formation towards NO. In this condition, despite the presence of active radicals, the maximum NOx removal efficiency will never be achieved (η_{NO} = 79% & η_{NOx} = 65%).

Research results indicate the fact that voltages above the corona threshold are not able to perform a complete NOx conversion (to N₂ and O₂) because of photochemical and light reactions at quantum level. Therefore, not only has no conversion been performed but the conditions for reverse conversion of NO₂ into NO have also been brought about [24, 25]. Researchers believe that by increasing the temperature, NO conversion will be greater than NOx [26]. Results of the present study also confirm this (Figs 5 and 9).

The comparison of NOx conversion process in plasma reactor (above 220°C) Figs 6 and 10 shows the interaction processes among influential factors of temperature, voltage, and reducer at a temperature of 220°C. As compared with the previous similar results, the conversion levels of NO and NO_x molecules decrease due to the burning of propane as a hydrocarbon at temperatures above 220°C; accordingly lack of free and active radicals for the reduction reaction and participation in plasma chemical reactions can be considered as a factor in the decline of NOx and NO conversion in this phase of experiments. A simultaneous increase in voltage (10 KV) and temperature intensifies the loss of conversion especially in the case of NO_x . Also, In addition, with reference to the results of previous studies, the generation of additional radicals, the imbalance between the reducer and the oxidant, and NO₂ partial reduction reactions in NOx matrix can affect the process in these conditions.

CONCLUSION

Under the condition of the present study, with a minimum arc generation, plasma reactor can remove maximum 79% of NOx inlet gases with 100 PPM concentration.

In order to initiate an electrical discharge in plasma environment, the corona initial voltage obtained is equal to 5KV at laboratory scale, under the optimal conditions of temperature and propane. This voltage is a function of cathode wire diameter, input current density, temperature, system relative pressure, and the plasma reactor geometry.

Results by NOx removal reveal that NO and NO₂ removal efficiency is proved to be optimal by a 50 HZ

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high voltage AC source at the voltage of 5KV, with propane molar ratio of 0.5, and the temperature of 180° C.

In this study, a minimal modification in the thermodynamic properties of the reactor inflow and the observance of the actual NOx emission limits have paved the way for implementing the project plan on a semi-industrial and industrial scale (specially for mobile sources).

Considering the conditions obtained through this study, the removal of NOx contamination in the air is considered as environmental clean technology due to the non-toxic gas outputs such as CO₂, H₂O, O₂, and N₂.

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