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Feasibility Study on the Removal of Nitric Oxide (NO) in Gas Phase using Dielectric Barrier Discharge Reactor

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ABSTRACT An initial study was carried out to observe the effect of dielectric barrier discharge on nitric oxide. A dielectric barrier discharge reactor unit consisting of a high voltage power supply and a coaxial dielectric barrier discharge tube was used. The discharge was observed to occur randomly, producing plasma with high energy particles. Technical air consisting of known concentration of nitric oxide, NO, was passed through the discharge. The NO concentration was measured continuously using an NO_x analyzer. This study shows that the removal rate of NO is highly dependent on the discharge voltage. Other factors influencing the removal rate is the gas flow rate and geometrical arrangement of the tubes.

ABSTRAK Satu kajian awal tentang kesan *'dielectric barrier discharge'* terhadap nitrik oksida telah dilakukan. Satu unit "dielectric barrier discharge" telah dibina menggunakan bekalan kuasa tinggi dan tiub nyahcas dielektrik sepaksi. Plasma yang dihasilkan adalah berbentuk rambang, mempunyai zarah bertenaga tinggi. Udara teknikal yang mengandungi nitrik oksida yang diketahui kepekatannya telah dialirkan melalui tiub dielectric tersebut. Kepekatan NO diukur menggunakan alat penganalisis NO_x. Kajian ini menunjukkan bahawa peratusan pengurangan banyak bergantung kepada voltan nyahcas tersebut. Faktor lain yang mempengaruhi kadar penghapusan adalah kadar aliran gas dan susunan geometric tiub-tiub tersebut.

(Dielectric barrier discharge, nitric oxide)

INTRODUCTION

Nitric oxide (NO) is a common gaseous pollutant that is harmful to mankind, vegetation and even materials. NO can be oxidised to become nitrogen dioxide (NO_2) which is also harmful and corrosive. When further exposed to sunlight, NO reacts with gaseous hydrocarbons and oxygen to form photochemical oxidants such as peroxyacetal nitrate (PAN) and ozone [1]. Exhaust gas from combustion fossil fuel such as coal and oil are one of the major contributors of toxic gaseous pollutants. In Malaysia, most of the nitrogen oxides are emitted from mobile and static sources.

There are two mechanisms where NO_x can be formed during combustion. The first is thermal NO_x where oxidation of N_2 occurs in the combustion air. The second is fuel NO_x that originates from the oxidation of chemically bound nitrogen in the fuel. NO_x may be removed by improving combustion process, such as using low NO_x burners, improve gas circulation, and staged combustion. It can also be removed using post combustion removal techniques such as the use of selective catalytic reduction or selective non catalytic reduction [1].

Studies on the removal of NO in flue gases using electron beam has been successfully carried out at the Malaysian Nuclear Agency as a collaborative project with TNB Research, a subsidiary of Tenaga Nasional [2]. This technique is based on the plasma oxidation of NO using accelerated electrons. Ionization caused by the impact of the accelerated electron causes the formation of gas phase radicals such as OH and HO_2. Subsequently, HNO_3 are formed through oxidation of NO by the radicals. At the end of the process, the HNO_3 is neutralized using ammonia to form ammonium nitrate. The nitrate occurs in fine powder form which can be collected using filter bags. Although the studies show that electron beam is effective in removing NO_x in flue gases, it involves high cost for actual implementation. This motivates researchers to look for more effective and economical method to remove this pollutant.

A dielectric barrier discharge reactor was developed at the Plasma Research Laboratory, University of Malaya. Initially it was used as an ozonizer to treat industrial wastewater [3]. In this study, the feasibility of using this dielectric barrier discharge for the removal of NO in gas phase is demonstrated.

MATERIALS AND METHODS

The dielectric barrier discharge (DBD) reactor consists of an AC high voltage power supply, 2 units of DBD cells and a flow meter. The DBD cell is made of coaxial electrodes that are insulated by a Pyrex test tube acting as the dielectric. A narrow space between the surface of the Pyrex tube and the outer earth electrode allows a stream of gas to flow through and it is within this narrow space that chemical reaction is expected to occur. When a 50 Hz AC high voltage of higher than 10 kV peak voltage is applied to the electrodes, micro-discharges will occur randomly between the surface of the Pyrex tube and the outer electrode across the narrow space. It is the micro-discharges which supply energy to the continuous stream of gas and hence inducing the reaction.

In this experiment, known concentrations of NO from a bottled source are used. The initial concentration is 51.2 ppm while the balance is a mixture of known concentration of SO_2 and nitrogen. The flow rate is controlled by the flow meter on the DBD reactor module. The dielectric discharge provides energetic electrons that produce radicals to react with the gas. A chemiluminescence's type NO analyzer (TEI Corporation, model 42C) is used to analyze the output continuously throughout the process. The removal efficiency is calculated by using the following equation:

$$\eta(\%) = \frac{[NO\,(in)] - [NO\,(out)]}{NO\,(in)} \times 100$$

Where η is the NO removal efficiency and the bracketed words denote whether the reading is from the inlet or the outlet.

In the first set of experiments, the DBD cells are connected such that the gas will flow through the two cells in parallel. The connection is later modified to allow the gas to flow inside the cells connected in series. Figure 1 shows the experimental setup used in both occasions. The discharge voltage however, remains the same for the two cases even though the flow configuration is different. The AC power supply is connected to both cells in parallel. The output voltage of the power supply is regulated using a variac. The experimental variables are the flow rates and the DBD cell discharge voltage. The flow rate is varied using the flow meter.

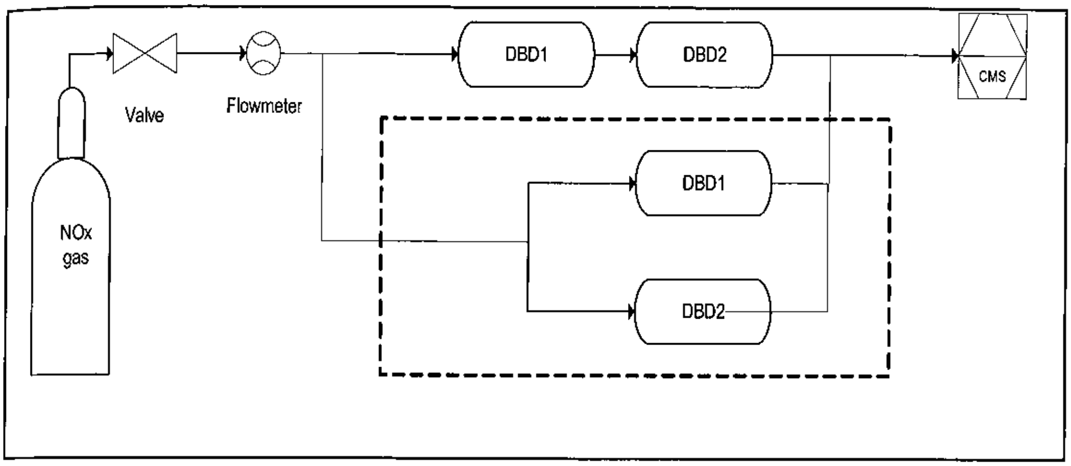

Figure 1. Schematic diagram for NO_x removal experimental setup. The DBD modules are connected either in series or in parallel (as shown in dotted-line box).

When investigating the effect of discharge voltage, the flow rate was fixed at 1 SCFH (standard cubic foot per hour) to control the consumption of the NO gas. The effective gas volume, i.e. the volume of gas that is actually in contact with the plasma, is calculated to be 0.06 litre. This gives the retention time, i.e. the amount of time the gas is in contact with the plasma to be 7.63 s. The voltage was later fixed at 28.2kV (the maximum energy at this stage) when running the experiment at different flow rates.

RESULTS AND DISCUSSION

Figures 2 and 3 show the effect of varying discharge voltage on the removal efficiency. Figure 2 shows the result obtained with parallel cell connection and Figure 3 shows the results for serial cell connection. Both figures show that the NO removal efficiency increases (thus the lower concentration) as the discharge voltage is increased. This is expected since at higher discharge voltage more radicals are produced to react with the NO.

The removal efficiency is found to be improved by connecting the two cells in series instead of parallel. The maximum efficiency for the case of parallel connection is 49% and it is increased to 63% for serial connection. The improvement is significant and the serial connection will be adopted for future work. By connecting the cells in series, the total processing rate may be reduced compared to parallel connection. This is also a factor that needs to be further investigated. Apparently the work with electron beam machines also gives similar findings. It was reported that two stages or double irradiation is better than single stage [2].

Varying flow rate is also found to affect the removal efficiency. This can be seen from the results shown in Figure 4. During the experiment, the flow rate cannot be varied over a wider range due to limitation on the reaction. At flow rate higher than 1.5 SCFH, there is no improvement in the removal efficiency.

At this point, it is reasonable to assume that most of the nitric oxide was reduced to nitrogen gas since oxygen is absent in the gas mixture. However, since SO_2 is also present and some reaction may cause O_2 to be dissociated and the O atom will subsequently react with the NO to produce NO_2 by oxidation. The possible reaction mechanisms are given below: [5], [6]

a. Formation of radicals
$e + N_2 \rightarrow N + N + e$
$e + O_2 \rightarrow O + O + e$
$e + H_2O \rightarrow OH + H + e$

b. The NO was removed by reduction reaction:
$NO + N \rightarrow N_2 + O$

c. Or by oxidation:
$NO + O \rightarrow NO_2$

Therefore, further investigations are needed to find the other products resulting from the reactions when the input is a mixture of several types of gases.

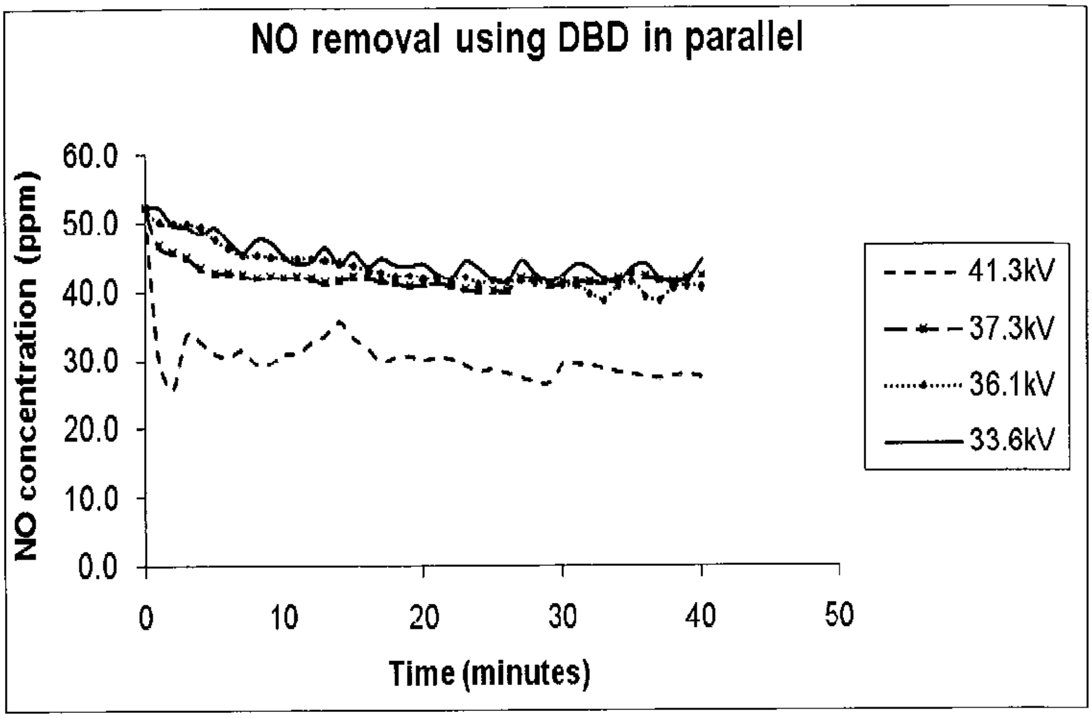

Figure 2. Removal of NO with flow connected in parallel.

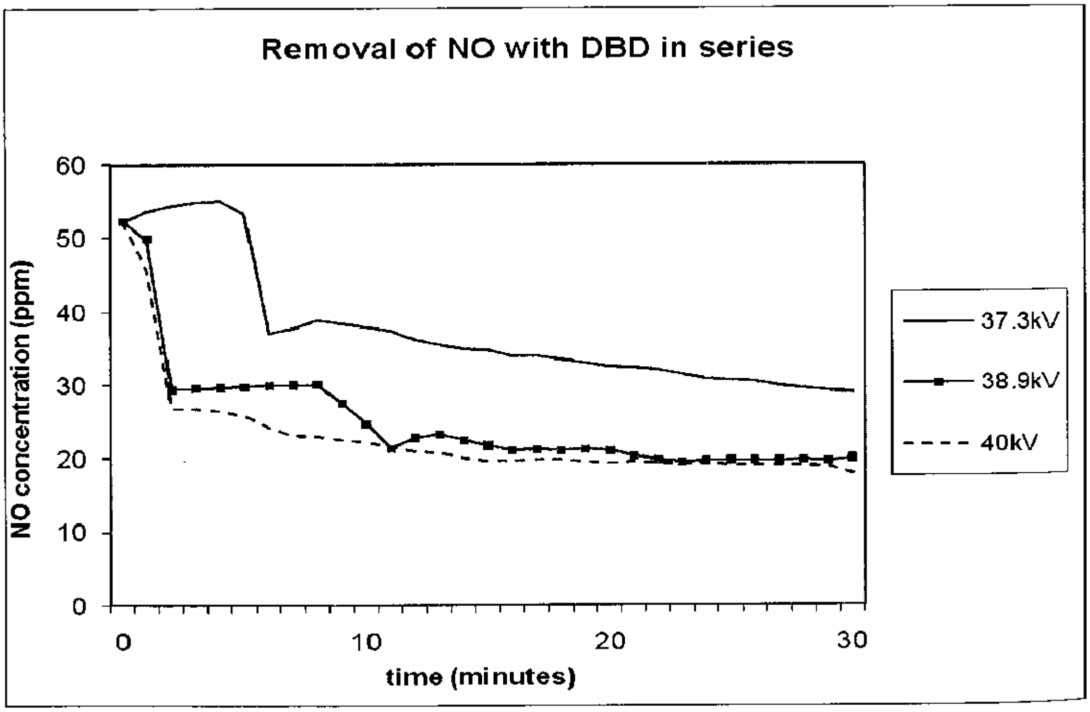

Figure 3. Removal of NO with flow connected in series.

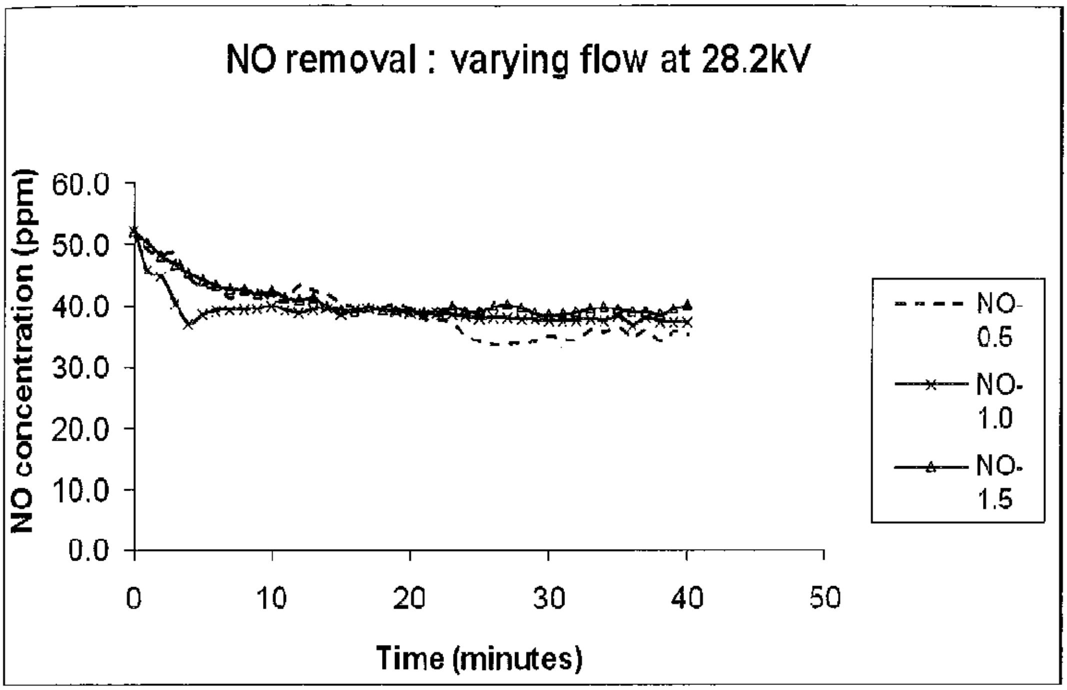

Figure 4. Removal of NO at different flow rate and fixed energy.

CONCLUSION

It has been demonstrated that the dielectric barrier discharge reactor is capable of producing similar effect as an electron beam machine to produce gas phase radicals to remove the NO. The removal efficiency is dependent on the flow rate and the discharge voltage of the dielectric barrier discharge cell. The flow configuration is also found to play an important role to improve the efficiency.

At this preliminary stage of the project, only the effects of flow rate, discharge voltage and geometrical arrangement of the dielectric barrier discharge tubes on the removal rates have been studied. Other aspects such as process temperature and humidity [7] will be examined in future experiments.

Potentially, the reactor is a versatile device to treat emission from various sources because of its modular design. Its adaptability depends on the number of DBD tubes or modules where the number of required modules will in turn depend on the flow rate of emission as well as the stack or exhaust pipe size.

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