

Production of Nitric Oxide Using a Pulsed Arc Discharge

Takao Namihira, *Member, IEEE*, Sunao Katsuki, *Member, IEEE*, Reuben Hackam, *Fellow, IEEE*, Hidenori Akiyama, *Fellow, IEEE*, and Kazufumi Okamoto

Abstract—Nitric monoxide (NO) is increasingly being used in medical applications. Currently, a gas cylinder of N₂ mixed with a high concentration of NO is used. This arrangement is potentially risky due to the possibility of accidental leak of NO from the cylinder. The presence of NO in air leads to the formation of nitric dioxide (NO₂), which is toxic to the lungs. Therefore, an on-site generation of NO would be very desirable for patients with acute respiratory distress syndrome and other related illnesses.

We have recently reported on the production of NO using a pulsed arc discharge. In the present work, the discharge reactor was made simpler and smaller. NO was generated using a pulsed arc discharge in dry air and in mixtures of oxygen and nitrogen. The composition of the gas mixture after treatment with an arc discharge followed by exposure to heated molybdenum was 540 ppm of NO, 48 ppm of NO₂, and the balance dry air at 0.1 MPa and 300 ± 3 K. No ozone was detected at the outlet of the system by UV absorption. The density of the brass particles emitted from the electrodes, which had diameters over 0.3 μm, was less than 1.39 μg/L. A filter could readily capture and thus remove the brass particles.

Index Terms—Acute respiratory distress syndrome (ARDS), endothelium-derived relaxing factor (EDRF), molybdenum converter, nitric oxide, pulsed arc discharge, pulsed power.

I. INTRODUCTION

FOR MEDICAL treatment, inhaled nitric oxide (iNO) has been used since NO was identified as an effective treatment involving endothelium-derived relaxing factor (EDRF) in 1987 [1]. Currently, iNO is being widely used as a cure for acute respiratory distress syndrome (ARDS), acute lung injury, persistent pulmonary hypertension of the newborn, and other related illnesses [2], [3]. iNO is also used in surgery and heart transplantation [4]. However, all current inhalation systems employ a gas cylinder of N₂ with a high concentration of NO. The concentration of NO in the gas cylinder is typically between about 500–800 ppm [5], [6]. An on-site generation of NO where it is used would be very desirable. In the present paper, the characteristics of a laboratory system for the production of NO which incorporates improvements to the design of the previous discharge reactor [7], [8] is reported.

II. EXPERIMENTAL SETUP AND PROCEDURE

Fig. 1 shows a schematic diagram of the system for generating NO. This system consisted of gas cylinders, a NO discharge reactor, a NO₂ converter, a gas analyzer, and gas pressure monitors. Gas cylinders of nitrogen (N₂), oxygen (O₂), and dry air were used. The proportion of O₂ in the mixtures of O₂ and N₂ was changed from 6% to 93%. The gas cylinder of dry air included 80% of N₂, 20% of O₂ and traces of carbon monoxide (<1 ppm), carbon dioxide (<1 ppm) and water vapor (<5 ppm). The gas inlet pressure (P_1) to the reactor was changed from 0.12 to 0.35 MPa. The gas outlet pressure (P_2) from the reactor was changed from 0.12 to 0.25 MPa. Both pressures (P_1 and P_2) were controlled using two valves, which were placed, respectively, at the inlet and outlet of the reactor. The difference between P_1 and P_2 was fixed at 0.1 MPa. The flow rate of the gas mixture and dry air were varied from 2.0 to 5.0 L/min at 273 K and 0.1 MPa using a mass flow controller (SEC-440J, ESTEC, Japan).

Fig. 2 shows the NO discharge reactor, a charging capacitor C (0.4 to 2.0 nF), a dc power supply (50 kV, 3 mA, HDV-50K3US, Pulse Electronic Engineering, Japan), a limiting resistor (2 MΩ), and a spark plug connected to a trigger pulse circuit. The trigger pulse circuit consisted of a capacitor (0.22 μF), a thyristor and a pulse transformer (1 : 200, HFT1009, Sanyo, Japan). This reactor was made simpler and smaller than that used previously [7], [8]. The outer diameter and the length of the reactor were 60 and 100 mm, respectively. The rod and the plate electrodes were made of brass. The brass rod ending with a hemisphere had a diameter of 10 mm. The distance between the rod and the plate was 5 mm. The charging voltage to the capacitor was increased from 15 to 30 kV with increasing outlet pressure (P_2) from the reactor. The charging voltage for all gas pressures was set at a level of about 80% of the self-breakdown of the gas. The pulse arc discharge between the rod and the plate was initiated by the UV radiation from the discharge between the spark plug and the plate electrode [9]–[13]. The pulse repetition rate was changed from 10 to 220 pps. The applied voltage to the rod electrode was measured using a voltage divider (EP-100K, Pulse Electronic Engineering, Japan), which was connected between the rod and the ground. The current to the reactor was measured using a Rogowski coil (Pearson current monitor, Model 110A, Pearson Electronics). The gas analyzer is based on the method of potentiostatic electrolysis [7], [14].

The NO₂ converter consisted of a stainless steel vessel enclosing a molybdenum (Mo) wire and a nickel–chrome wire heater. The molybdenum wire diameter was 0.1 mm and had a

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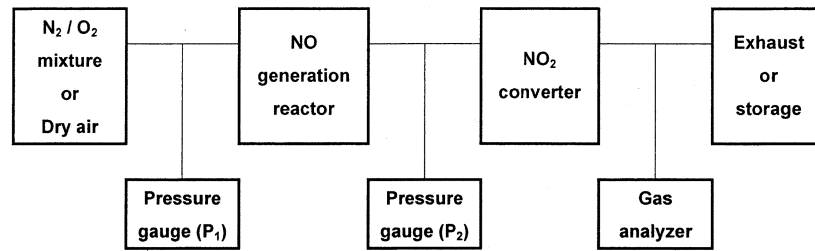


Fig. 1. Schematic diagram of the experimental setup.

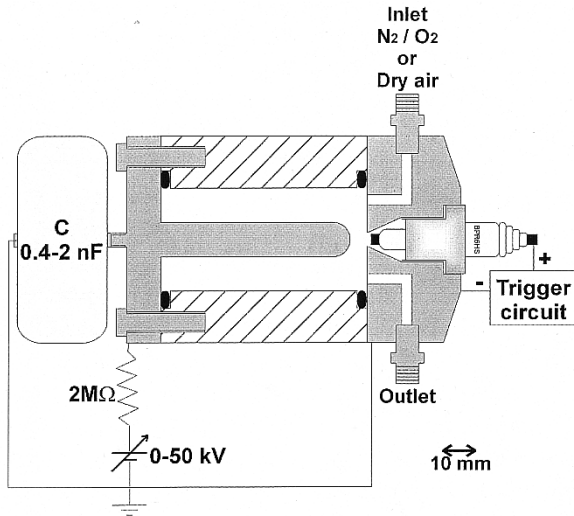
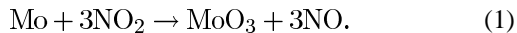


Fig. 2. NO discharge reactor and electrical circuit.

length of 100 m. The vessel was heated to about 670 K. Above 600 K, NO_2 is converted to NO through the following reaction [7], [15]:



The performance of the NO_2 converter was studied in a subsidiary experiment using NO_2 mixed with either N_2 or dry air obtained from gas cylinders. The concentration of NO_2 in N_2 and in dry air was changed from 36 to 225 ppm at 2 L/min.

An assessment of the NO production system including the NO_2 converter was performed. The concentration of O_3 was measured using an UV absorption measurement (Ubest V-570DS, JASCO, Japan) at the same location of the gas analyzer (Fig. 1). The concentration of the brass particles in the gas mixture produced by the pulsed arc discharge was determined by passing the gas through a Whatman silica filter ($0.3 \mu\text{m}$) with a combination of a sensitive electronic balance (10^{-3} g, MJ-300, YMC Co., Japan). Table I shows the conditions of all experiments.

III. RESULTS AND DISCUSSIONS

A. Applied Voltage to and Discharge Current in the Reactor

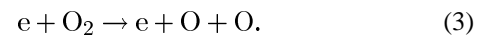
The pulse arc discharge between the rod and the plate electrodes occurred when a trigger voltage having a peak of 14 kV was applied to the spark plug. Typically, it took about $0.45 \mu\text{s}$ for the discharge current to fully develop after application of the trigger pulse. Both voltage and current waveforms showed

damped oscillations. The oscillation frequency decreased from about 10 to about 5 MHz when the external capacitance C was changed from 0.4 to 2.0 nF. This is consistent with $\omega^2 = 1/LC_t$, where ω is the radian frequency, L the total inductance, and C_t the total capacitance which includes that of the external circuit and the discharge.

Fig. 3 shows the dependence of the resistance (R) of the arc plasma on time after its initiation for different external capacitances and, therefore, different input energies. The resistance of the arc plasma was determined using the attenuation constant (γ) on the positive slopes of the current waveforms. The attenuation constant is expressed by $\gamma = R/2L$ [10]. The resistance of the arc plasma increased with decreasing capacitance and this is attributed to lower arc current. In the case of 2 nF, the temperature of the plasma was shown to be about 10^4 K [7]. The resistance of the plasma, for example, at 2 nF (Fig. 3) had good agreement with other data [16].

B. NO and NO_2 Concentrations

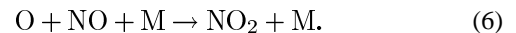
Following the application of HV pulsed power to a mixture of oxygen and nitrogen, the radicals N and O are created by electron impact processes [17], [18]



These radicals are largely responsible for the creation of NO [19]



NO is removed from the plasma by oxidation with the radical O [17], [20]



M is a third molecule such as N_2 or O_2 . NO_2 is also produced by [21], [22]

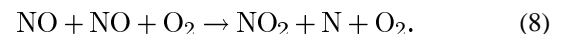
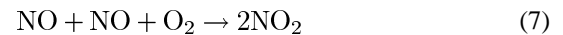


Fig. 4 shows the concentrations of NO and NO_2 as a function of the proportion of O_2 in the mixture of O_2 and N_2 . It can be seen from Fig. 4 that the concentrations of both NO and NO_2 initially increased with increasing proportion of O_2 in the mixture. This is because the production of NO requires the presence

TABLE I
EXPERIMENTAL CONDITIONS

	Proportion of O ₂ in the mixture of O ₂ and N ₂ [%]	Concentration of NO ₂ [ppm]	Inlet pressure to reactor (P ₁) [atm]	Outlet pressure from reactor (P ₂) [atm]	Charging voltage to capacitor [kV]	Gas flow rate (reduced 273 K and 1 atm) [L/min]	Capacitance of capacitor [nF] C	Pulse repetition rate [pps]	Temperature of vessel [K]
Expt. #1	6-93 (Mixture)	-	2.2	1.2	15	2.0	2.0	30	300
Expt. #2	20 (Dry air)	-	2.2-3.5	1.2-2.5	15-30	2.0	2.0	20	300
Expt. #3	20 (Dry air)	-	2.2	1.2	15	2.0-5.0	2.0	50	300
Expt. #4	20 (Dry air)	-	2.2	1.2	15	2.0	0.4-2.0	10-220	300
Expt. #5	0 (Nitrogen)	36-223	2.2	1.2	-	2.0	-	-	670
Expt. #6	20 (Dry air)	39-225	2.2	1.2	-	2.0	-	-	670
Expt. #7	20 (Dry air)	-	2.2	1.2	15	2.0	0.4	220	670

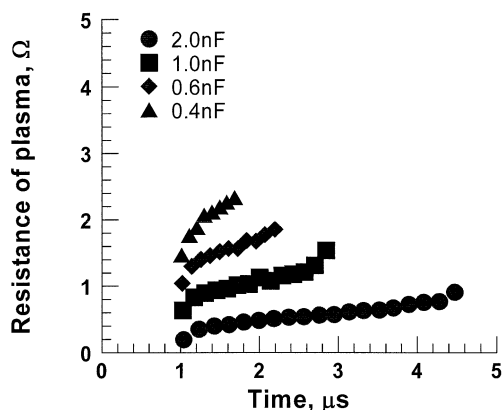


Fig. 3. Resistance of the arc discharge as a function of time after its initiation. Charging voltage 15 kV. Other conditions: Expt. #4 of Table I.

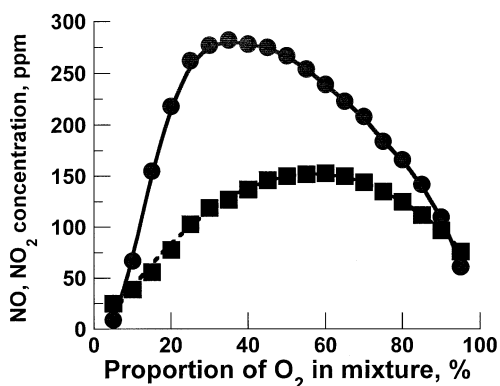


Fig. 4. Concentrations of NO and NO₂ as a function of the percentage of O₂ in a mixture of O₂ and N₂. Conditions: Expt. #1 of Table I.

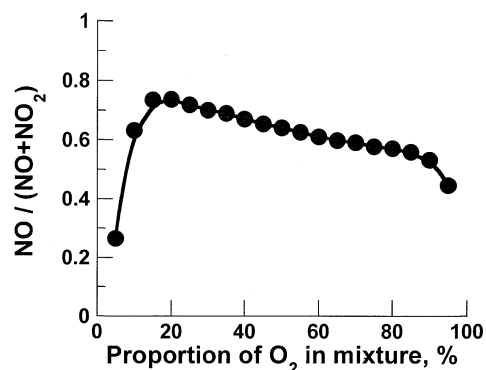


Fig. 5. Ratio of NO/(NO+NO₂) versus percentage of O₂ in a mixture of O₂ and N₂. Conditions: As in Fig. 4.

of oxygen [(3)–(5)]. The concentration of NO reached a maximum for 35%–50% of O₂ in the mixture. As the proportion of O₂ was increased >50%, the concentration of NO started to decrease because of the reduced concentration of N₂ and N [(2), (4), and (5)]. The concentration of NO₂ increased and reached a maximum at 50%–70% of O₂ in the mixture. This was while NO concentration was decreasing (Fig. 4) which is consistent with equations (6)–(8) due to conversion of NO to NO₂. The maximum concentrations of NO and NO₂ reached 282 and 153 ppm, respectively.

Fig. 5 shows the ratio of NO/(NO+NO₂) as a function of the proportion of O₂ in the mixture of O₂ and N₂. It is desirable that the ratio of NO/(NO+NO₂) should have a high value because NO₂ is toxic to humans. The ratio of NO/(NO+NO₂) had a peak, which had value of 0.74 when the proportion of O₂ in the mixture of O₂ and N₂ was 20% (Fig. 5). At this mixture proportion the production of NO can be made in dry air. Therefore, this choice of gas mixture would be most economical.

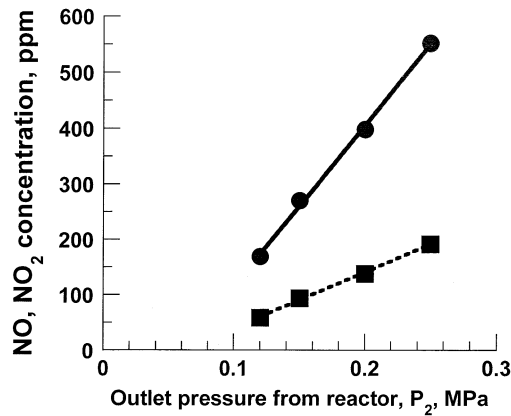


Fig. 6. Concentrations of NO and NO₂ versus P_2 . Conditions: Expt. #2 of Table I. ● NO; ■ NO₂.

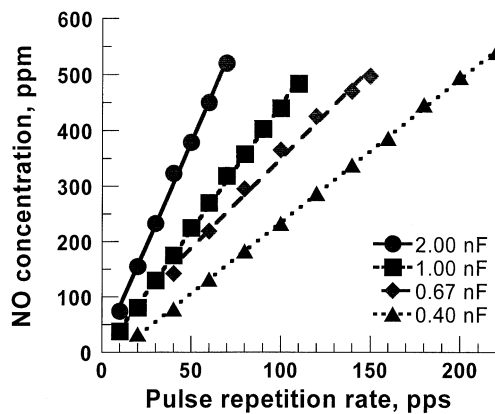


Fig. 7. Dependence of the concentrations of NO on pulse repetition rate. Conditions: Expt. #4 of Table I. ● 2.0; ■ 1.0; ◆ 0.67; and ▲ 0.4 nF.

Fig. 6 shows the concentrations of NO and NO₂ as a function of the exhaust pressure (P_2) in dry air. The concentrations of NO and NO₂ increased with increasing the outlet pressure. This is due to increasing charging voltage and, therefore, increasing input energy to the discharge. The ratio of NO/(NO+NO₂) had a constant value of 0.74 in the range of 0.12–0.25 MPa of outlet pressure.

The concentrations of NO and NO₂ decreased with increasing the gas flow rate due to decreasing residence time of the gas in the reactor. Typically, the concentration of NO decreased from 379 to 160 ppm and that of NO₂ from 119 to 59 ppm when the flow rate increased from 2 to 5 L/min. The ratio of NO/(NO+NO₂) decreased slightly from 0.76 to 0.73 with increasing the gas flow rate in the same range.

Fig. 7 shows the concentrations of NO as a function of the pulse repetition rate for different capacitances. The concentrations of NO (Fig. 7) and NO₂ (not shown for brevity) increased linearly with increasing pulse rate due to increased energy input to the discharge. Likewise, the concentrations of NO and NO₂ increased with increasing capacitance (higher input energy) at a fixed pulse rate. Fig. 8 shows the ratio of NO/(NO+NO₂) as a function of the pulse repetition rate for different capacitances. This ratio initially increased with increasing capacitance and pulse repetition rate and, therefore, with increasing energy input into the discharge, until it saturated at 0.77.

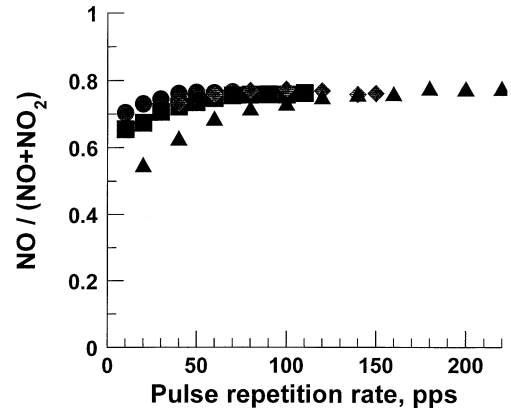


Fig. 8. Dependence of NO/(NO+NO₂) on the pulse repetition rate for different charging capacitance. Conditions: Expt. #4 of Table I. ● 2.0; ■ 1.0; ◆ 0.67; and ▲ 0.4 nF.

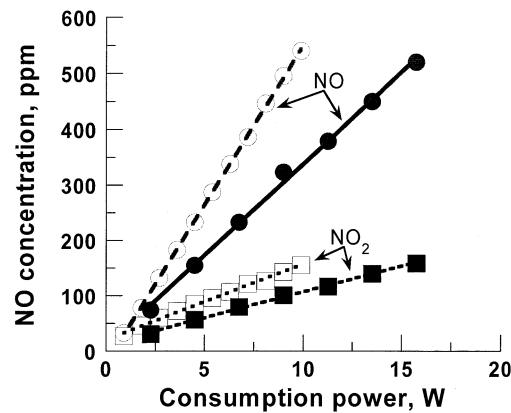


Fig. 9. Dependence of NO and NO₂ concentrations on average dc power consumption for different capacitance. Conditions: Expt. #4 of Table I. Close symbol: 2.0; and open symbol: 0.4 nF.

Fig. 9 shows the dependence of the concentrations of NO and NO₂ on the consumption of average power of the dc supply for different capacitances. The consumption power (P , in Watts) was obtained from

$$P = \frac{1}{2} \times C \times V^2 \times f \quad (9)$$

where V and f are the charging voltage (in volts) to the capacitor and the pulse repetition rate [pps].

It will be observed from Fig. 9 that for a given concentration of NO, the power requirement decreased with decreasing capacitance. This could be due to faster rise time of the discharge current. Therefore, a smaller capacitance led to higher energy efficiency for NO production system. The ratio of NO/(NO+NO₂) increased with increasing the consumption power from about 0.55 at 0.9 W to 0.77 at 7.0 W and higher.

C. Characteristics of NO₂ Converter

Fig. 10 shows the dependence of the concentrations of NO and NO₂ on time of exposure to heated Mo in nitrogen. The initial concentration of NO was 28 and of NO₂ was 223 ppm. It will be seen from Fig. 10 that NO increased with time due to the decomposition of NO₂. In nitrogen, the concentration of NO₂ decreased to 0 ppm while in dry air to 16 ppm (not shown). The

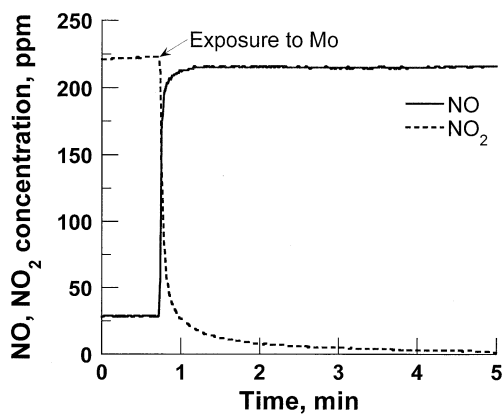


Fig. 10. Dependence of NO and NO₂ concentrations on time of exposure to heated Mo. Conditions: Expt. #5 of Table I. Initial concentrations: NO, 28; NO₂, 223 ppm; and balance, N₂.

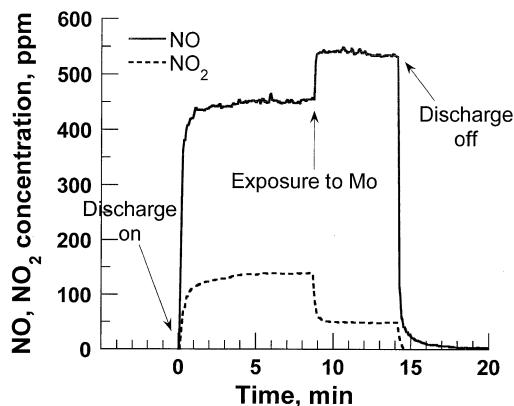


Fig. 11. Dependence of the concentrations of NO and NO₂ on time of application of pulsed power in dry air including the flow of the gas over heated Mo. Conditions: Expt. #7 of Table I.

concentration of NO increased to 216 ppm both in nitrogen and in air. The final concentration of NO₂ increased linearly with increasing initial concentration of NO₂ in dry air (Expt #6 of Table I) after exposure to heated Mo. In nitrogen, the concentration of NO₂ decreased to 0 ppm for all initial concentrations of NO₂ in the range 36 to 223 ppm, while in dry air it decreased to 18 ppm for the highest initial concentration.

The conversion ratios were 1.0 and 0.9, respectively, in N₂ and in dry air over a wide range of initial concentration of NO₂ from 36 to 225 ppm. The conversion ratio is defined as

$$\text{Conversion ratio} = \frac{\text{NO}_{2i} - \text{NO}_{2f}}{\text{NO}_{2i}} \quad (10)$$

where NO_{2i} and NO_{2f} are, respectively, the initial and the final concentrations of NO₂ after exposure of the gas mixture to heated Mo.

D. Assessment of the NO Reactor System

Fig. 11 shows the dependence of the concentrations of NO and NO₂ on time subsequent to the application of the pulsed power to the reactor in dry air including the passage of the gas mixture over a heated Mo. The conditions of this experiment were chosen to yield the highest concentration and the

highest energy efficiency for the production of NO. It will be observed from Fig. 11 that the concentrations of NO and NO₂ increased rapidly following the starting of the pulsed discharge and soon became constant. In the next step, the gas was passed over a heated molybdenum and the concentration of NO increased while that of NO₂ decreased. The concentrations of NO increased from 455 to 540 and that of NO₂ decreased from 138 to 48 ppm. In this case, the conversion ratio from NO₂ to NO was 0.65. The concentration of O₃ and brass particle having diameters over 0.3 μm were zero and less than 1.39 μg/L, respectively, measured at the location of the gas analyzer. The brass particles could be removed using a filter. The final ratio of NO/(NO+NO₂) was 0.92.

IV. CONCLUSION

A discharge reactor system to produce NO using a pulsed arc discharge was studied. The following are the main results.

- 1) The NO reactor system was more compact than earlier systems.
- 2) The concentration of NO was controlled by adjusting the outlet pressure of the gas mixture, the gas flow rate, and the discharge energy (the charging capacitance and the pulse repetition rate).
- 3) Ozone was not detected at the outlet of the system.
- 4) The highest concentration of NO obtained was 540 ppm.
- 5) The lowest concentrations of NO₂ obtained after passing the gas over a Mo converter were 0 in N₂ and 48 ppm in dry air while NO was 540 ppm.

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