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Production of Nitric Monoxide Using Pulsed Discharges for a Medical Application

Takao Namihira, Shunsuke Tsukamoto, Douyan Wang, Sunao Katsuki, Reuben Hackam, Fellow, IEEE, Kazufumi Okamoto, and Hidenori Akiyama, Senior Member, IEEE

Abstract—Nitric monoxide (NO) is widely used in medical treatment of acute respiratory distress syndrome (ARDS). The production of NO is of interest to the medical community. In the present work, NO is generated by pulsed discharges between two rod electrodes in a mixture of nitrogen and oxygen. An arc discharge having a temperature of about 10 000 K was produced, which was sufficient to generate NO. Some of the important parameters affecting the production of NO have been investigated. These include the percentage of O2 (6–94%) in the mixture of N2 and O2, the energy of the discharge (0.5–12 J/pulse), the pulse repetition rate (0.5–4.5 pps) and the flow rate (1.35–5.4 l/min) of the gas mixture. NO2 produced in the discharge was successfully changed to NO using a heated molybdenum tube. NO2 must be extracted from the gas before clinical inhalation. The concentration of ozone was completely eliminated by bubbling the gas mixture through water. A maximum of NO and a minimum of ozone were generated when the proportion of O2 in the gas mixture was in the range of 20–27%. The concentrations of NO and NO2 increased with increasing pulse repetition rate and with decreasing flow rate of the mixture. In all cases, NO2 was effectively removed using a heated molybdenum tube.

Index Terms—Acute respiratory distress syndrome, medical application, nitric monoxide, pulsed arc discharge.

I. INTRODUCTION

RECENTLY, Nitric monoxide (NO), which is known to be a toxic gas, has been used in the medical community because it has a relaxing effect on the blood vessels [1]. For example, a disease of the lung that includes the acute respiratory distress syndrome (ARDS) has a high death rate of about 50%. It is being cured by inhalation of NO (iNO) [2],[3]. Fig. 1 shows a patient under medical treatment for ARDS with all accessory equipment including the gas analyzer which detects NO and NO2, the NO gas cylinder, the mixing of O2 and air, the humidifier and the water trap [4]. This is a complex arrangement and a more suitable and safe device to inhale NO is required. Many doctors are currently using an industrial cylinder of NO to provide iNO [5]. However, an industrial cylinder might be risky if a leak is present, since it would generate NO2 by mixing NO with O2. The toxic effects to the lungs of NO2 at high concentrations are well known [4]. Therefore, a device to ensure a safe inhalation of NO is needed for many patients who have a lung disease. A production of NO using a gliding arc was reported by Richard et al. [6]. However, dc applied voltage was employed and only air at atmospheric pressure was considered.

In the present work, the purpose is to develop a safe system for the generation and inhalation of NO. A pulsed arc discharge was used to generate NO. A pulsed arc discharge has many advantages compared to dc arc. They are as follows:

1) the plasma temperature is higher than using dc;
2) the damage to the electrodes is smaller than with dc;
3) the pulsed discharge is more stable.

In the present experiment, the generation of NO using pulsed arc discharge was confirmed. The gas composition of O2 and N2, the energy of the pulse, the pulse repetition rate, and the gas flow rate have been varied in order to obtain the most suitable concentration of NO for medical applications. The pulsed arc discharge generates NO2 with NO. Because a small quantity of NO2 is very toxic for the patient [7], NO2 is removed using a heated molybdenum tube.

II. EXPERIMENTAL SETUP AND PROCEDURE

Fig. 2 shows the experimental setup. The electrical circuit consisted of a dc power supply (50 kV, 0.3 mA), the charging resistance, the capacitor, spark gap switch (SGS) and a pair of brass rods as the electrodes. The brass rods had diameters of 12 mm and their ends were at a radius of curvature of 5 mm. The distance between the electrodes was 5 mm. At first, the power supply charged the capacitor. The SGS was closed by a trigger
pulse (15 kV), and a pulsed arc discharge developed between the electrodes. These conditions were common to 2.1 and 2.2.

A. Determination of the Plasma Temperature of the Pulsed Arc Discharge

The light emissions from the pulsed arc discharge were measured by an optical multichannel analyzer (OMA, Atagobussan, Japan). The energy of the pulse was changed by changing the capacitance from 2 to 36 nF. The dc charging voltage was constant at 25 kV. The temperature of the pulsed arc plasma was calculated using the line pair method [8]. This method uses the relative ratio of emission intensities on the plural spectrum lines. The temperature of the plasma is derived from the following equation:

\[
\ln \left( \frac{I_\lambda}{A g_u} \right) = -\frac{1}{kT} E_u + \ln C
\]

where \( I_\lambda, \lambda, A, g_u, E_u, k, T, \) and \( C \) are the relative ratio of emission intensity, the wavelength, the transition probability, the statistical weight of the upper level, the energy of the upper level of the copper lines, Boltzmann’s constant, the temperature of plasma and a constant for various atoms, respectively. The transition probabilities, the statistical weights of the upper level and the energy of the upper levels of the copper lines are shown in Table I [9].

Assuming a linear plot of \( \ln(I_\lambda/A g_u) \) versus \( E_u, T \) can be calculated. The discharge electrodes were made of brass which includes copper. The four copper lines were used as the plural spectra lines. They have the wavelengths of 510.55, 515.32, 521.82, 529.25 nm. Table I shows \( A, g_u, \) and \( E_u \) for these four lines.

B. Conditions for NO Production

In Fig. 2, a molybdenum tube (internal diameter, 4.6 mm; length, 100 mm) was inserted between the chamber housing the electrodes and the gas analyzer to serve as NO\(_2\) converter. A mixture of nitrogen and oxygen at \( 1.01 \times 10^5 \) Pa was used at the input to the arc discharge chamber. The concentrations of NO and NO\(_2\) were measured using the gas analyzer after a steady state was reached (within 5 min). The gas analyzer is based on the method of potentiostatic electrolysis [10]–[12]. This method relies on the characteristic reaction of oxidation and de-oxidation of the surface of an electrode immersed in an electrolyte solution. The current which is measured at a constant applied voltage gives a measure of the gas concentration. Different voltage is applied for measuring NO and NO\(_2\). In the present work, the electrode was made of gold and the electrolyte was sulfuric acid. The instrument (testo 33) is calibrated by the manufacturer (Hodakastest, Japan).

The proportion of O\(_2\) in the mixture of O\(_2\) and N\(_2\) was changed from 6 to 93%. The energy of the pulse was changed by changing the capacitance from 2 to 36 nF. The charging voltage was constant at 25 kV. The pulse repetition rate was changed from 0.5 to 4.5 pps (pulses per second). The flow rate was changed from 0.9 to 5.4 l/min. The temperature of molybdenum was changed from 298 to 873K in order to study the effectiveness of molybdenum on the removal of NO\(_2\). A stainless steel tube, having the same dimensions as for the molybdenum, was also used but found to be ineffective. Table II shows the conditions of the experiment. In each column only one parameter is varied while all others remained constant. Five different experiments, as shown in Table II, were performed.
III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Characteristics of the Pulsed Arc Discharge

Fig. 3 shows typical waveforms of the applied voltage and the discharge current between the electrodes. Both waveforms oscillated after applying the trigger voltage. Fig. 4(a) and (b) show the four copper spectra lines using 2 and 36 nF, respectively. These four copper lines were obtained in all capacitance values (2–36 nF).

Fig. 5 shows a plot of \( \ln(\frac{I_{\lambda}}{Ag_{\lambda}}) \) versus \( E_u \) for different applied energy (\( C = 2 \) to 36 nF). The straight-line approximation using (1) is used to get an approximate gas temperature, even though the data points deviate from the straight line. It can be seen from Fig. 5 that the straight lines for different capacitors are of the same slope. The gas temperature was found to be about 10 000 K. This temperature is sufficient to generate NO, since the mixture of N\(_2\) and O\(_2\) is transformed to NO at gas temperatures above 1000 K.

B. Characteristics of NO Production

Fig. 6 shows the concentrations (in parts per million, ppm) of NO, NO\(_2\), and the ratio of NO\(_2\)/(NO + NO\(_2\)) as a function of \( O_2/(O_2 + N_2) \times 100 \), under the experimental conditions of expt. #1 in Table II.

Fig. 7 shows the dependence of the concentrations of NO, NO\(_2\), and NO\(_x\) on the capacitance and therefore on the energy of the pulse (charging voltage was constant at 25 kV). The concentrations of NO and NO\(_2\) increased with
increasing the value of the capacitance and therefore of the energy of the pulse. The value of NO$_2$/NO$_X$ decreased with increasing the value of capacitance up to 20 nF and thereafter remained constant. Fig. 8 shows the concentrations of NO, NO$_2$, and NO$_2$/NO$_X$ as function of the pulse repetition rate. The concentrations of NO and NO$_2$ are in direct proportion to the repetition rate. Concurrently, the value of NO$_2$/NO$_X$ was constant at each repetition rate. The measurement at 0 pps (Fig. 8) was taken when high voltage pulses were not applied to the mixture of N$_2$ and O$_2$. Fig. 9 shows the concentrations of NO, NO$_2$, and NO$_2$/NO$_X$ as function of the gas flow rate. The concentrations of NO and NO$_2$ decreased with increasing the gas flow rate. Concurrently, the value of NO$_2$/NO$_X$ was constant at 0.25 in the range 1.35 to 5.40 l/min.

Fig. 10(a) shows the concentrations of NO, NO$_2$, and NO$_2$/NO$_X$ as function of the temperature of molybdenum. Fig. 10(b) shows the concentrations of NO, NO$_2$, and NO$_2$/NO$_X$ as function of the temperature of stainless steel. In the case of molybdenum, the concentration of NO increased and that of NO$_2$ decreased with increasing the temperature of molybdenum starting from about 720 K. However, in the case of stainless steel, NO and NO$_2$ concentrations were constant in the range 298–873 K [Fig. 10(b)]. It is evident that the concentrations of NO increased and that of NO$_2$ decreased due to the presence of molybdenum. With molybdenum, NO$_2$ is converted to NO because the decrease in NO$_2$ matched the increase in NO. NO$_2$ was converted according to the reaction [13]

$$3\text{NO}_2 + \text{Mo} \rightarrow 3\text{NO} + \text{MoO}_3,$$  

(2)

The concentration of ozone which is produced in the arc discharge was measured with the instrument available in the laboratory (Ebara Jitsugyo co., model DOA300). This instrument relies on the absorption of ultraviolet (UV) at 235.7 nm where
the absorption cross section is highest. For conditions necessary to obtain the suitable concentrations of NO for medical applications (proportion of O₂ in the mixture = 26%, capacitance = 20 nF, pulse repetition rate = 4.5 pps, gas flow rate = 2.0 l/min and charging dc voltage = 22 kV), the concentration of ozone was largely removed using heated Mo at 870 K. With a temperature of about 10 000 K, the concentration of ozone can be completely eliminated by bubbling the mixture through water. (0.5 l/min) are necessary.

4) The concentration of ozone can be completely eliminated by bubbling the gas mixture through water.

The concentration of NO. This was done and the concentration of ozone was found to decrease from 10 to 0 ppm.

IV. CONCLUSION

Production of nitric monoxide using pulsed discharges was studied for medical application. The results are summarized as follows:

1) NO was produced using pulsed arc discharge at a gas temperature of about 10 000 K.

2) NO₂ was largely removed using heated Mo at 870 K.

3) To obtain the highest concentration of NO, the proportion of O₂ of about 26%, the capacitance value as large as possible (36 nF), the pulse repetition rate as high as possible (4.5 pps) and the gas flow rate as low as possible (0.5 l/min) are necessary.

4) The concentration of ozone can be completely eliminated by bubbling the gas mixture through water.

The productions of NO using air, and the detail measurements of gas composition following discharges therein are the subject of a future study. This method of production of NO using pulsed arc discharge in air will be tested using animals.

REFERENCES


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