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Study of ozone generation in an atmospheric dielectric barrier discharge reactor

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ABSTRACT

Ozone (O_3) generation in a dielectric barrier discharge (DBD) reactor driven by a pulsed power supply was investigated at atmospheric pressure and room temperature. An O_3 generation efficiency model is established in which discharge power, O_2 concentration, gas flow rate, and volume of the discharge space are included. Constants in the O_3 generation efficiency model were obtained by fitting the model with experiment results. O_3 concentration can be simply calculated from the energy density and initial O_2 concentration. Comparison on O_3 concentrations from calculation with references is given.

Introduction

Ozone (O_3) is a useful chemical and widely used in many fields, such as advanced oxidation processes (AOPs), chemical–biological processes (CBP), and semiconductor industry [1,2]. O_3 also is a powerful chemical in food and medical treatments [3–7]. Generally, O_3 is generated by applying high-voltage to a dielectric barrier discharge (DBD) reactor of a discharge space in which an oxygen (O_2)-containing gas is present. It has been understood that a number of tiny breakdown channels occur in the discharge space; those channels are suggested as microdischarges having a time order of microseconds, where O_3 is generated [8–18]. O_3 generation reactions in microdischarges begin with the dissociation of O_2 molecules to oxygen atoms (O) by the impact of O_2 with energized electrons in an electric field. O atoms then combine with O_2 to yield O_3. The energy efficiency of O_3 generation is strongly related with the production efficiency of O atoms in the microdischarges.

The energy efficiency of O_3 generation (ξ) using an AC power supply can be obtained from the approximation given by Eliasson and Kogelschatz [8], as defined by

$$\xi = \frac{2\rho_D}{e v_d E/n}. \tag{1}$$

Wei et al. [19] developed a numerical model which describes the influence of both electrical and discharge configuration parameters on ozone concentration in pulsed positive dielectric barrier discharge. Factors of pulse repetition frequency, difference of peak pulsed voltage and corona inception voltage, gap length, relative permittivity, gas pressure, gas flow rate, and pulse duration were taken into account. O_3 concentration is given in a form in which 9 parameters are required.

Related with the dissociation of O_2 to O by the impact of O_2 with electrons as shown in Eq. (2), the constant k_1 of the dissociation process is sensitive to the amplitude of the electric field. The value of k_1 in $1/(cm^3\,s)$ can be calculated using Eq. (3), when a microwave power supply is applied [20].

$$e + O_2 \xrightarrow{k_1} 2O + e, \tag{2}$$

$$k_1 = 2 \times 10^{-\left(7.8 + \frac{14.7}{\theta}\right)} + 10^{-\left(7.4 + \frac{17.1}{\theta}\right)}, \tag{3}$$

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Nomenclature

Symbols	
d	alumina spacer thickness in mm
E	energy injection over one pulse discharge duration in J/Hz
E_0	electric field amplitude in V/cm
ED	energy density in J/m^3
f	pulse frequency in Hz
F	total gas flow rate at the inlet of the DBD reactor in m^3/s
ΔF	difference on total gas flow rates between the inlet and outlet of the DBD reactor in m^3/s
E/n	reduced electric field in Td
I_i	discharge current in A at discharge time t_i
I_{i+1}	discharge current in A at discharge time t_{i+1}
k	ozone generation rate constant in $mol^{0.5}\ m^{1.5}/J$
k_1	kinetic constant rate of O_2 dissociation in $1/(cm^3\ s)$
N_m	concentration of neutrals in $1/cm^3$
P	energy injection density in W/m^3
P_{in}	energy injection power in W
P_{INC}	inception energy injection power for O_3 generation in W
r_{O_2}	O_2 consumption rate in mol/J
r_{O_3}	O_3 generation efficiency in mol/J
T_e	temperature of electrons in eV
t_i	discharge time in s
t_{i+1}	discharge time in s
V_i	discharge voltage in V at discharge time t_i
V_{i+1}	discharge voltage in V at discharge time t_{i+1}
V_R	total discharge space volume in m^3
x	O_2 conversion in percentage
α	constant
β	constant
η	O_3 generation efficiency in g/kWh
θ	energy related factor in eV
v_d	electron drift velocity in cm/s
ν_e	collision frequency of electrons with neutrals in Hz
ξ	number of oxygen atoms produced per eV in 1/eV
ρ_D	total O_2 dissociation rate coefficient in cm^3/s
ω	frequency of the microwave field in Hz
$[O_2]_0$	initial O_2 concentration in inlet gases of the DBD reactor in mol/m^3
$[O_2]$	O_2 concentration in mol/m^3
$[O_3]$	O_3 concentration in g/m^3
$[O_3]'$	O_3 concentration in mol/m^3
*	excited state
Abbreviations	
CT	current transformer
DBD	dielectric barrier discharge
e	electron
HV	high-voltage
OSC	oscilloscope

$$\theta = \frac{E_0 \cdot \nu_e}{\left(\omega^2 + \nu_e^2\right)^{\frac{1}{2}} N_m} \times 10^{16}. \tag{4}$$

k_1 was also suggested as a constant of 2×10^{-9} $1/(cm^3\ s)$ [21]. k_1 can be obtained by Lee et al. using Eq. (5) in a DBD reactor [22]. k_1 is given in a more complicated form in which parameters, such as the energy branch to electron, excitation rate, impact power, discharge area, channel height of gas flow path, and average electron density, are required [23]; such parameters are difficult to be obtained. However, the influence of energy on ozone generation is additionally required if k_1 is given in a constant form.

$$k_1 = 4.2 \times 10^{-9} \exp\left(-\frac{5.6}{T_e}\right). \tag{5}$$

Recently, ozone generation using DBD reactors is still an active study [24–28]. The aim of this work is to find an O_3 generation efficiency model in which factors or parameters are simply obtained from experiments. At first, the O_3 generation in a DBD reactor driven by a pulse power supply was experimentally investigated. Secondly, an O_3 generation efficiency model was established using factors of discharge power (energy injection), O_2 concentration, gas flow rate, and volume of the discharge space those are easily given or measured. Finally, constants in the O_3 generation efficiency model were obtained after fitting the model with experiment results.

Experimental setup

Fig. 1 shows the experimental setup for the investigation of O_3 generation in a DBD reactor. A pulsed high-voltage (HV) from a power supply (DP-12K5-SCR, PECC, Japan) was applied to the DBD reactor. The discharge voltage and current waveforms were measured using a voltage probe (V-P, P6015A, bandwidth DC–75 MHz, Tektronix, USA) and a current transformer (CT, TCP0030, bandwidth DC–120 MHz, Tektronix, USA), respectively. The signals from the voltage probe and current transformer were digitized and recorded using a digital phosphor oscilloscope (OSC, DPO 3034, bandwidth 300 MHz, Tektronix, USA).

The DBD reactor consists of two alumina plates (purity 96%, 50 × 50 × 1 mm^3, Kyocera, Japan) and two metal electrodes (aluminum tapes, 31 × 31 mm^2), those plates and electrodes were sandwiched closely. The distance between two alumina plates was adjusted using two alumina spacers of different thicknesses ($d = 0.29$, 0.85, 1.48, 2.03, or 2.36 mm). Microdischarges occur in a discharge space between two alumina plates when high-voltage is applied to two metal electrodes. A gas mixture of nitrogen (N_2, purity 99.9999%) and O_2 (purity 99.9999%) was supplied to the inlet of the DBD reactor using two mass flow controllers (MFC) at a fixed

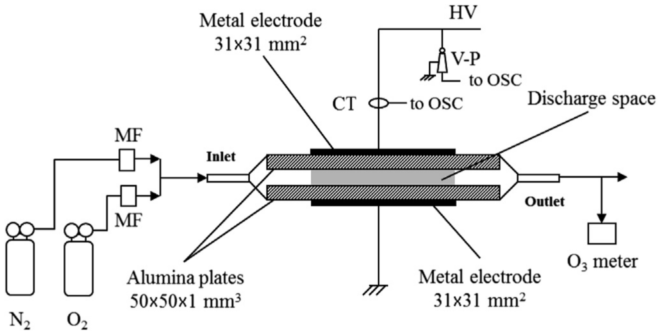

Fig. 1. Schematic diagram of O_3 generation in a DBD reactor.