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論　文

Optimum Operating Conditions for Ozone Generation in AC Corona and AC Silent-discharge Reactors

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The linear relationships between the specific energy and the energy density (or Becker parameter) are investigated for ozone generation by AC corona and AC silent-discharge. These relationships are used to determine the ozone formation and destruction coefficients in dry-air-fed wire-cylinder AC-corona and silent-discharge reactors. These coefficients depend on the investigated parameters of the reactor; namely, the radius of the discharge wire, the air flow rate and the active length of the reactor. The ratio between the specific energy and the ozone concentration, referred to as the economical criterion parameter, is assessed as a function of the energy density for the different investigated parameters. The optimum operating conditions for ozone generation can be estimated by the minimum values in the economical criterion parameter proposed in this study. This condition is valid both for the AC corona and AC silent-discharges until the saturation of the ozone concentration.

1. Introduction

Among the methods for generating non-equilibrium plasmas at atmospheric pressure and temperature are the AC corona and the AC-silent discharge [1)].

There are numerous applications of AC silent discharges. Typical examples include the elimination of NO_x and SO_x from flue gases [2-4)], pumping CO_2 lasers and excimer lamps, surface treatment and modifications as well as CO_2 hydrogenation [5)]. On the other hand, AC corona generates light, audible noise, radio noise and ozone [6)] around the stressed conductor. AC corona produces also bipolar ions used for discharging charged dielectric plates by neutralizing their charges [7,8)]. AC corona on EHV lines is used to attenuate the over voltage traveling waves along these lines [9)], thus reliefing the electric stress on the line insulation.

The characteristics of corona and silent discharges as influenced by the geometry of the discharge electrode have been investigated before by the same authors [10)]. The corona discharges have been generated in wire-cylinder and wire-duct reactors stressed by dc and ac voltages. The pulse characteristics of silent discharges are compared with those of ac corona discharges. The current-voltage and the ozone generation characteristics of these discharges are recorded. These characteristics depend significantly on the geometry of the reactor irrespective of the discharge type.

Ozone being generated by AC corona and AC silent discharge has many applications in industries. These include deNOx and deSOx processes[2-4)], water purification without ojfectionable byproducts and residues[3,4)], oxidation of oleic acid and synthesis of certain hormones, vitamins and some perfumes[11)]. The ozone layer, which is apparently endangered by anthopogenic emissions, protects the biosphere against dangerous short-wave radiations from the sun [12)].

A global approach was first proposed by Becker and Lunt [13-15)] from the electrosynthesis of some chemical products (ozone). Global rate coefficients K_f and K_d were introduced to describe respectively the ozone formation and destruction phenomena in discharge reactors. These kinetic rates are generally considered as proportional to the energy density [16)].

Experimental measurements resulting from ozone electrosynthesis pointed out a fundamental parameter $\beta=P/Q$, relating the discharge power input into the reactor P with the gas flow rate Q, assuming homogeneous energy distribution in the flowing gas inside the reactor [13-15)]. This is well known as the Becker (or energy density) β parameter and is acknowledged as an important dynamic characteristic of the reactor. The gas flow rate Q is equivalent to the volume of the reactor $V_{reactor}$ swept by the running out gas during the residence time t_r ($Q=V_{reactor}/t_r$). Both $v_{reactor}$ and t_r depends on the geometrical characteristics of the reactor.

In relation to the Becker parameter, the specific energy ε is defined as representing the energy necessary to obtain one molecule of product per unit volume; $\varepsilon=\beta/C$, where C is the volumetric concentration of the resulting product (ozone) inside the flowing gas. The optimum product concentration inside the reacting carrier gas corresponds to the minimal energy consumption in the reactor. Thus, it is possible to define an economical criterion related parameter γ as equal to the ratio between the energy expended for one molecule of product (the energy density) and the product concentration C: $\gamma=\varepsilon/C=\beta/C^2$. The minimum value of γ dictates the reactor operating conditions for optimum ozone generation.

Key words: Ozone, Ozonizer, Non-thermal plasma, Silent discharge, Corona discharge,

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Empirical equations have been proposed [14] to account for ozone synthesis in silent discharges. These equations relate the Becker parameter β, the formation and destruction factors K_f and K_d to (i) the ozone volumetric concentration C, (ii) the specific energy ε, (iii) the parameter γ and (iv) the conditions for optimal operation of the discharge reactor.

This paper is aimed at investigating how applicable these equations for ozone generation by AC corona in comparison with AC silent discharge. This calls for experimental measurement of the concentration of the ozone generated in AC corona and AC silent discharges reactors. Wire-cylinder reactors fed by dry-air are selected in this work. Among the goals of the paper is also to seek an avenue to optimize the ozone generation in these reactors.

2. Experimental set-up and Technique

2.1 Experimental set-up

Figure 1 shows a schematic diagram of the experimental set-up used in this study. It is composed of the high voltage power supply, the discharge reactor and the gas flow system.

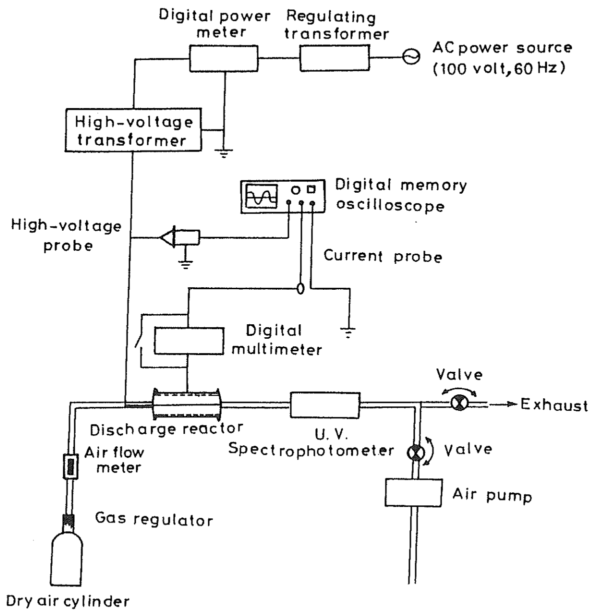

Fig. 1 Schematic diagram of the experimental setup.

2.1.1 High-voltage power supply

It is a setup transformer rated 0.1/20 kV sinusoidal, 60Hz and 1 kVA. The input electric power was adjusted with a regulating transformer (500 VA) and measured with a digital power meter (Yokogawa 2534). The transformer output voltage applied to the discharge wire of the reactor was displayed on a two channel digital real time memory oscilloscope (Tektronix TDS360P, 200 MHz) by using a 1000:1 high-voltage probe (Tektronix P601S) with a band width of 100 MHz.

2.1.2 Discharge reactors

A coaxial wire-cylinder reactor was used in this study. For AC corona, the outer cylinder was made from aluminum with inner diameter of 2 cm and thickness of 1 mm. The edges of the cylinder were curved to avoid local discharges. For AC silent discharge, the outer cylinder was an aluminum foil of 0.3 mm thickness wound on the external surface of a glass tube of 1.76 cm inner diameter and 1.2 mm thickness. The aluminum cylinder or foil was grounded. The discharge wire was tensioned axially in the cylinder and the wire radius was varied in the range 0.025-0.2 mm. The active length L of the reactor (which goes in discharge) was variable in the range 10-20 cm.

2.1.3 Gas flow system

The reactor was fed by dry air from a high- pressure cylinder connected to it through an air-flow meter. The ozone concentration was determined from absorption measurements using a quartz cell attached to a UV-1200 spectrophotometer operated at a wavelength of 253.7 nm, close to the maximum of Hartley absorption band of ozone [17]. The air flow rate Q was measured at atmospheric pressure and temperature which varied between 20° and 21° C. The flow rate Q was variable in the range 0.5-8 L/min.

2.2 Measuring technique

The discharge reactor under study was connected to both the high-voltage power supply and the gas flow system. Details are reported elsewhere [10,18].

3. Results and Discussions

The specific energy (ε= β/C) of ozone generation by AC corona and AC silent-discharge generated in dry air-fed wire-cylinder reactors is calculated as a function of the investigated parameters (discharge wire radius r, flow rate Q and reactor length L).

All the ε= f(β) relationships of the AC silent discharge and some of these relationships of the AC corona fit straight line equations in the range of the density up to the beginning of the ozone saturation concentration. This is shown in Fig.2 for different values of the discharge wire radius. These relationships are shown for different flow rates Q, Fig.3, and different values of the reactor length L, Fig.4. In general, this conforms with the Becker relation, which leads to linear expression of the specific energy (ε) as a function of the energy density (β) as reported in Table 1. Therefore, the linear equations describing the ε=f(β) relationships determine the ozone formation and destruction coefficients, K_f and K_d. The K_f values have been determined from the intersection of the straight lines with the ε-axis (the ordinates equal $1/K_f$ when β tends to zero). The slope of the straight lines is $[(K_f+K_d)/K_f]$, which determines K_d values.

Tables 2 - 4 summarize the K_f and K_d values of the ozone generated in dry air-fed wire-cylinder reactors for different operating conditions.

In order to judge the existence of optimal operating conditions for ozone generation by AC corona and AC silent discharge in dry air-fed wire-cylinder reactors, the economical criterion parameter (γ) has been calculated and plotted as a function of (β) as shown in Figs. 5-7. These figures show that most of the γ=f(β) curves have minimum positions which corresponds to the condition δγ/δβ=0. The optimal operating conditions for ozone generation by AC corona and AC silent discharge in dry air-fed reactors have been determined corresponding to (γ_{min}) over the whole range of the investigated parameters.

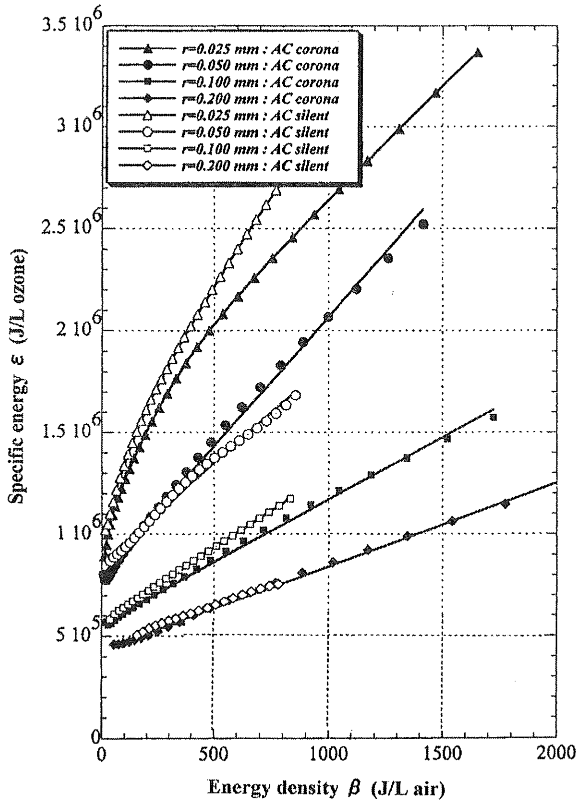

Fig 2 Specific energy (ε) as a function of energy density (β). (L=15cm and Q=1 L/min)

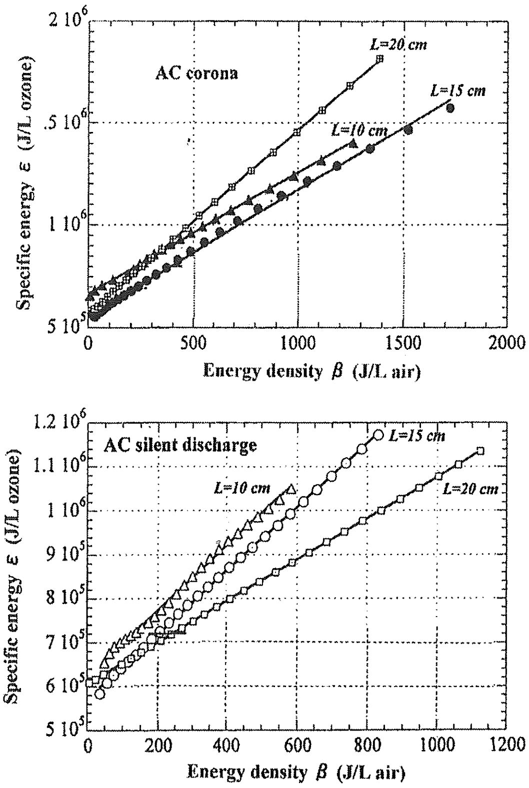

Fig. 4 Specific energy (ε) as a function of energy density (β). (r=0.1 mm and Q =1 L/min)

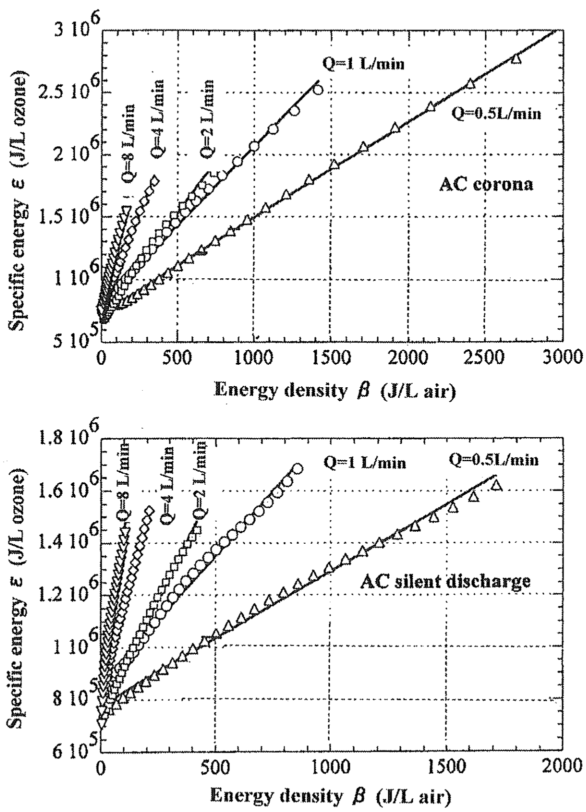

Fig. 3 Specific energy (ε) as a function of energy density (β). (L=15 cm and r=0.05 mm)

Table 1 Ozone production characteristic quantities.

Type of the Flowing gas	Air
Ozone concentration	The Becker relation $C = \dfrac{K_f \beta}{1+(K_f + K_d)\beta}$
Specific energy	$\varepsilon = \dfrac{1}{K_f} + \dfrac{(K_f + K_d)}{K_f}\beta$
Economical criterion	$\gamma = \dfrac{[1+(K_f + K_d)\beta]^2}{K_f^2 \beta}$
The optimal operating conditions; $(\delta\gamma/\delta\beta)=0$	$\beta_{\min} = 1/(K_f + K_d)$ $C_{\min} = 0.5K_f/(K_f + K_d)$ $\varepsilon_{\min} = 2/K_f$ $\gamma_{\min} = 4(K_f + K_d)/K_f^2$

Table 2 The optimum operating conditions for ozone generation for different values of wire radius r. (L=15cm and Q=1 L/min)

Type	r(mm)	K_fx10^{-6} (l/J)	K_d x10^{-3} (l/J)
AC corona	0.050	1.254	1.5940
	0.100	1.784	1.0883
	0.200	2.312	0.9453
AC silent discharge	0.05	1.127	1.1566
	0.100	1.748	1.294
	0.200	2.206	0.8756

Table 3 The optimum operating conditions for ozone generation for different air flow rates Q. (L=15 cm and r=0.05 mm)

Type	Q(L/min))	K_fx10^{-6} (l/J)	K_d x10^{-3} (l/J)
AC corona	0.5	1.373	1.0559
	1	1.254	1.5937
	2	1.470	2.4959
	4	1.316	4.0577
	8	1.323	6.7019
AC silent discharge	0.5	1.285	0.6586
	1	1.167	1.1566
	2	1.351	2.3877
	4	1.236	4.2836
	8	1.271	8.3107

Table 4 The optimum operating conditions for zone generation for different values of the reactor length L. (r=0.1 mm and Q=1 L/min)

Type	L(cm)	K_fx10^{-6} (l/J)	K_d x10^{-3} (l/J)
AC corona	10	1.496	0.8789
	15	1.784	1.0883
	20	1.750	1.5622
AC silent discharge	10	1.599	1.1816
	15	1.748	1.2694
	20	1.652	0.7781

The values of β_{min} (corresponding to γ_{min}) obtained from Figs.5-7 for AC corona agreed reasonably with those calculated from the relationship $\beta_{min} = (1/K_f + K_d)$ of Table 1 (corresponding to the optimal operating conditions) as depicted in Tables 5a-5c.With the knowledge of β_{min}, the minimum power P_{min} (=β_{min} Q) fed to a given reactor for a given flow rate Q is determined. This defines the optimum operating conditions for this reactor at that flow rate.

The overall ozone formation coefficient (K_f) increases with the increase of the discharge wire radius for AC corona reactors, the same as for AC silent reactors as shown in Table 2.

Table 5 (a) Values of β_{min} (J/L) for AC corona as determined by the discharge wire radius. (L=15 cm, Q=1 L/min)

Wire radius (mm)	β_{min} (from Fig.4)	β_{min} =1/(K_f+ K_d)
0.05	706	699.515
0.10	1190	1183.94

Table 5 (b) Values of β_{min} (J/L) for AC corona as determined by the air flow rate. (L=15 cm, r= 0.05 mm)

Flow rate (L/min)	β_{min} (from Fig.5)	β_{min} =1/(K_f+ K_d)
0.5	830	842.984
1.0	706	699.515

Table 5 (c) Values of β_{min} (J/L) for AC corona as determined by the active length of the reactor. (r= 0.1 mm, Q=1 L/min)

Reactor length (cm)	β_{min} (from Fig.6)	β_{min} =1/(K_f+ K_d)
15	1190	1183.94
20	697	688.083

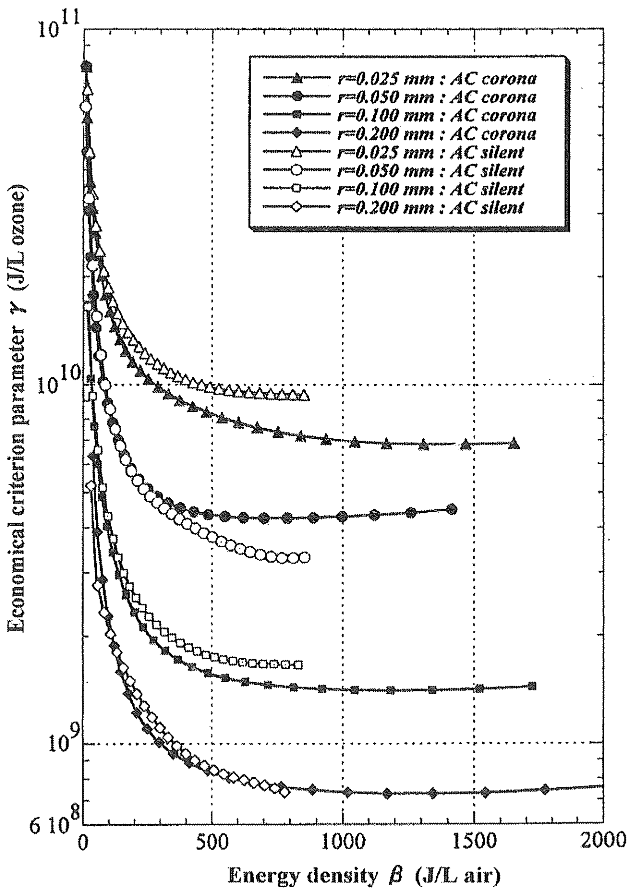

Fig. 5 Economical criterion parameter (γ) as a function of energy density (β). (L=15cm and Q=1 L/min)

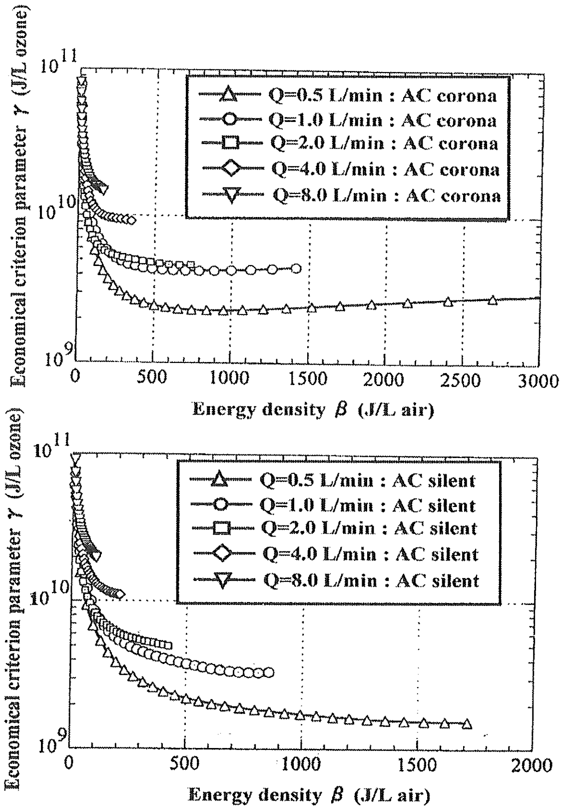

Fig. 6 Economical criterion parameter (γ) as a function of energy density (β). (L=15cm and r=0.1mm)

The overall ozone destruction coefficient (K_d) mostly increases with the increase of the gas flow rate inside the AC corona reactors, the same as for AC silent reactors as shown in Table 3.

At a constant flow rate of Q=1 liter/minute, the overall ozone formation coefficient (K_f) in dry air-fed wire-cylinder reactors reaches its maximum value at a reactor length of L=15cm for AC corona, the same as for AC silent discharge as shown in Table (4).

The K_f and K_d values in Tables (2)-(4) can be used with the Becker relation for assessment of ozone generation in the investigated reactors for the same operating conditions in range of the energy density (β) up to beginning of the ozone saturation concentration. The Becker relation cannot be used to explain such saturation concentration of the generated ozone [19].

The ozone concentration generated in wire-cylinder reactors increases with the increase of the stressed wire radius for the same discharge power applied to the reactors whatever the discharge type (corona or silent discharge). The larger the discharge wire radius, the higher is the concentration of the generated ozone at the same operating conditions. This tendency can be explained by the fact that the volume of the ionization-zone around the stressed wire, where the discharge plasma is confined, increases with the increase of the discharge wire radius. This conforms with previous studies [20-22]. The increase of the plasma thickness around the discharge wire means an increase of the path length for the electron collision with the flowing gas molecules inside the plasma layer, with a subsequent dissociated oxygen atoms required for the ozone generation.

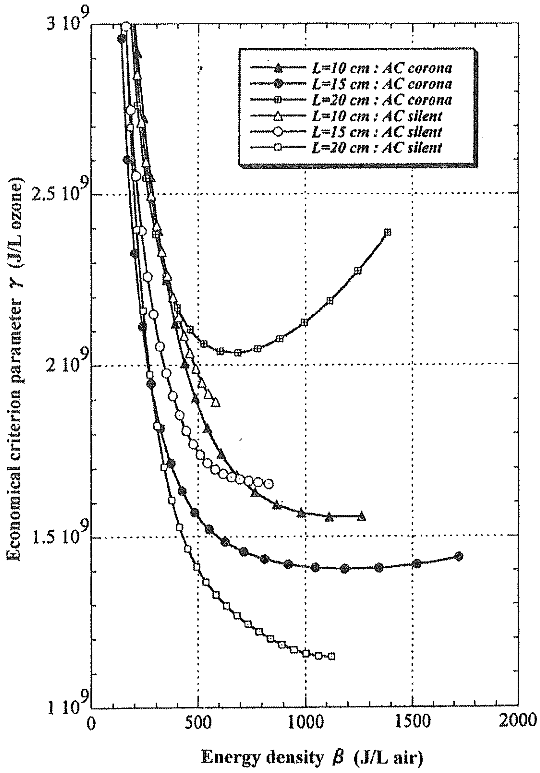

Fig. 7. Economical criterion parameter (γ) as a function of energy density (β). (r=0.1mm and Q=1 L/min)

This is why the specific energy increases with the decrease of the wire radius for the same energy density, Fig.2.

The concentration of ozone generated in the investigated reactors decreases with the increase of the gas flow rate inside the reactors for the same discharge power applied to the reactors whatever the discharge power (corona or silent discharge). The faster the gas flow rate the lower is the concentration of the generated ozone at the same operating conditions. This is explained by the decrease of residence time of the gas molecules within the ionization-zone as the gas flow rate increases inside the reactors, where the ozone is generated [23-25]. This is why the specific energy increases with the increase of the gas flow rate for the same energy density, Fig.3.

With the increase of the reactor length, the residence time of the flowing air molecules increases within the discharge plasma. This is in addition to the enhancement of the thermal distribution between the gas and the reactor walls [25]. Subsequently, the effect of the increase of the reactor length on the ozone generation characteristics is the same as that of the decrease of the gas flow rate. This is why the specific energy increases with the decrease of the reactor length for the same energy density, Fig4.

All of this explain why the ε=f(β) relationships of AC coronas generated in dry air-fed reactors fit linear relations in range of β only up to beginning of the ozone saturation concentration, the same as for AC silent discharges.

4. Conclusions

The linear relationship between the specific energy and the energy density (or Becker) parameter is verified for ozone generation by AC corona, the same as for AC silent discharges.

(1) This relationship holds for dry-air fed wire-cylinder reactor with different values of the investigated parameters; namely, the radius of discharge wire, the air flow-rate and the active length of the reactor.

(2) The optimum operating conditions for ozone generation can be estimated by the minimum values in the economical criterion parameter proposed in this study. This condition is valid both for the AC corona and AC silent-discharges until the saturation of the ozone concentration.

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