

[54] **CORONA REACTION METHOD AND APPARATUS**

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Related U.S. Patent Documents

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[57] **ABSTRACT**

Corona induced chemical reactions are conducted in a corona discharge zone in which narrow high voltage pulses are applied along with a relatively low voltage bias potential. It is found that for many corona discharge reactions, such as the conversion of oxygen to ozone, the present method increases the electrical efficiency of the reaction.

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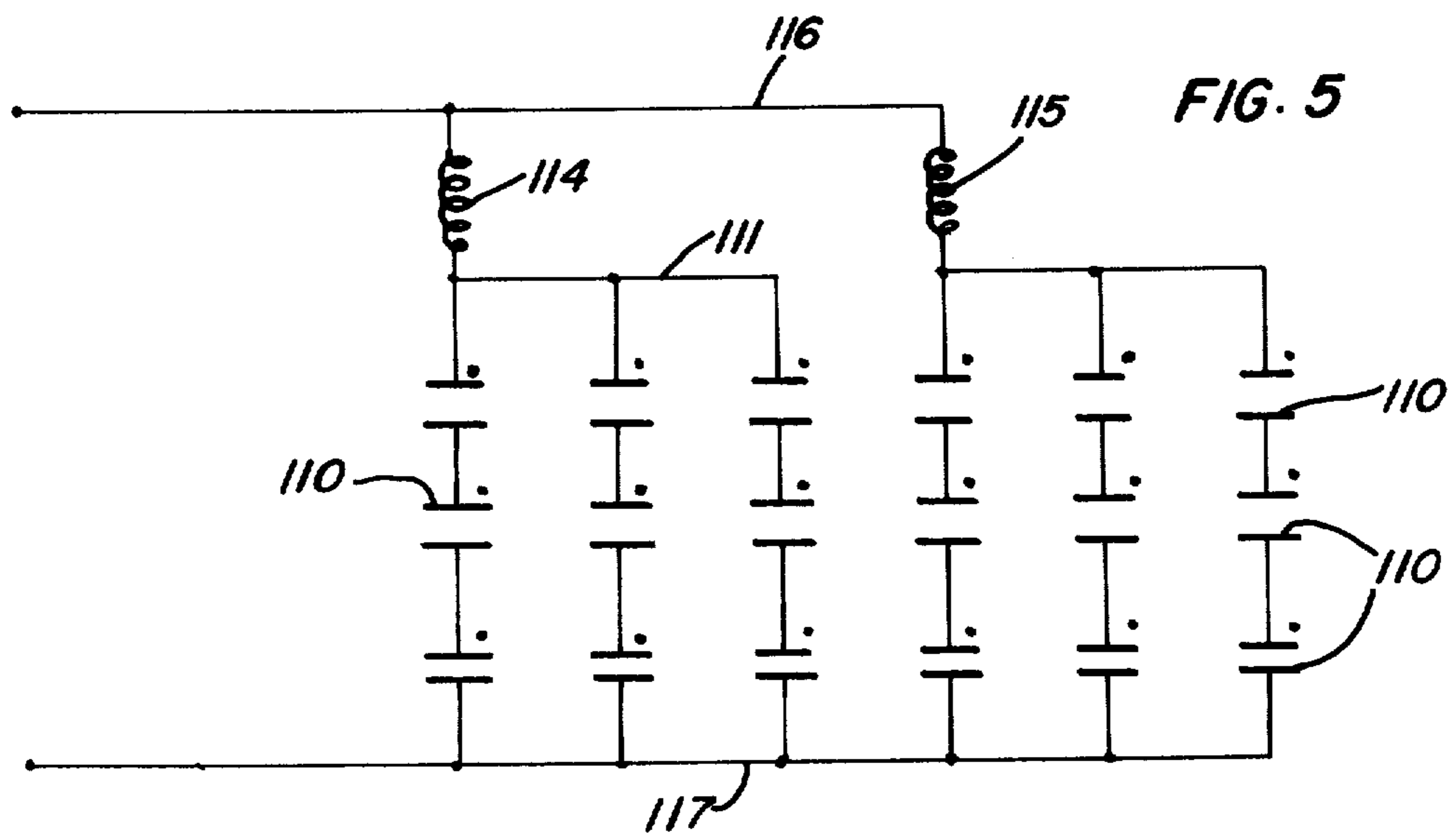
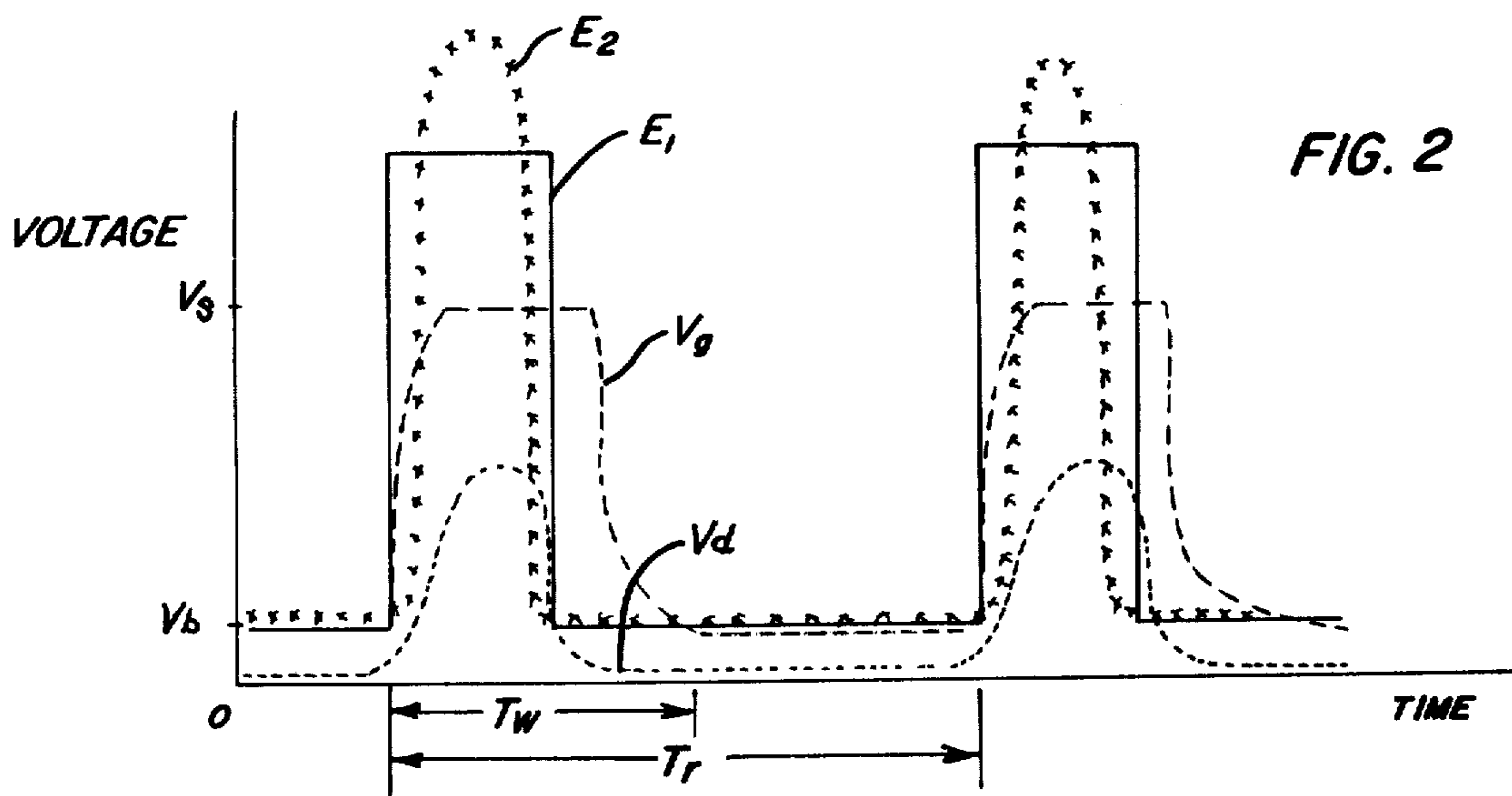
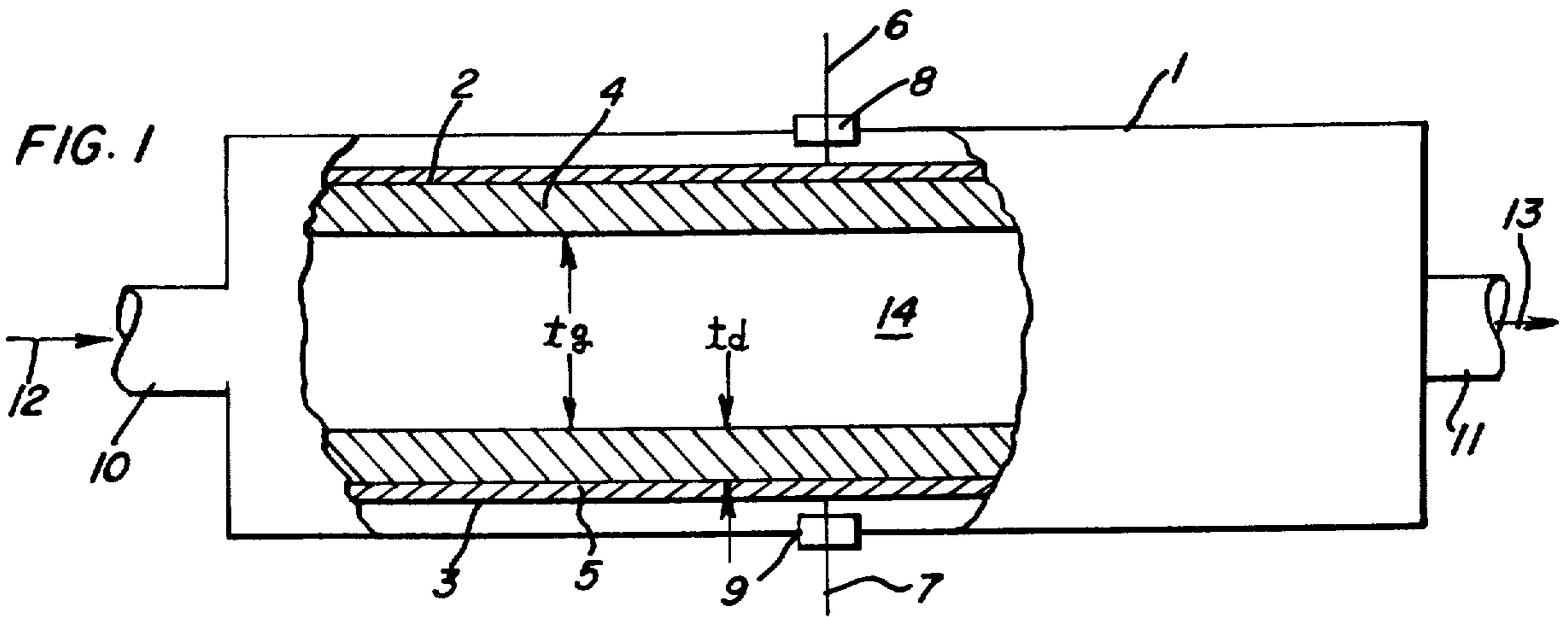
[58] Field of Search **204/176; 250/532-541**

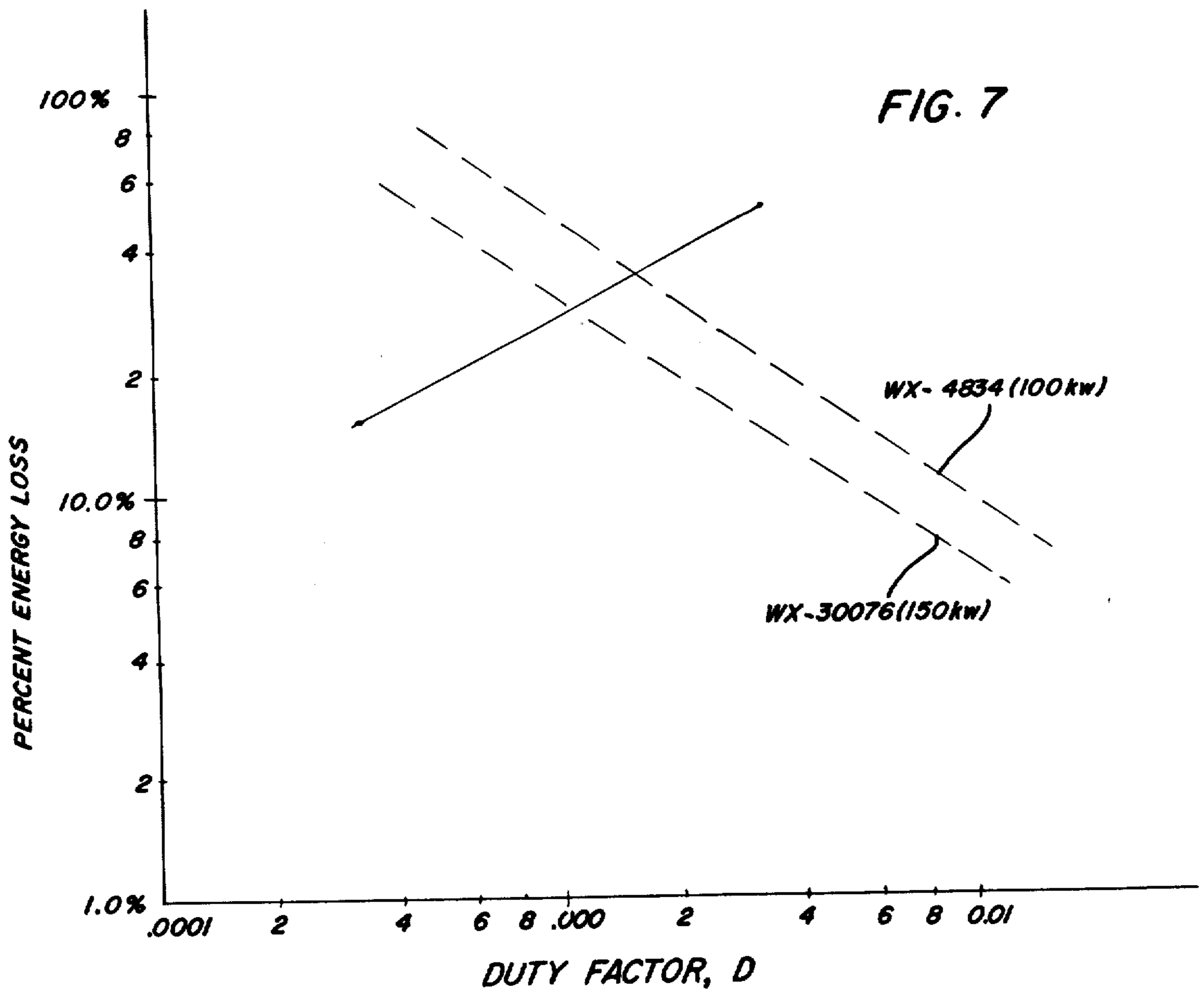
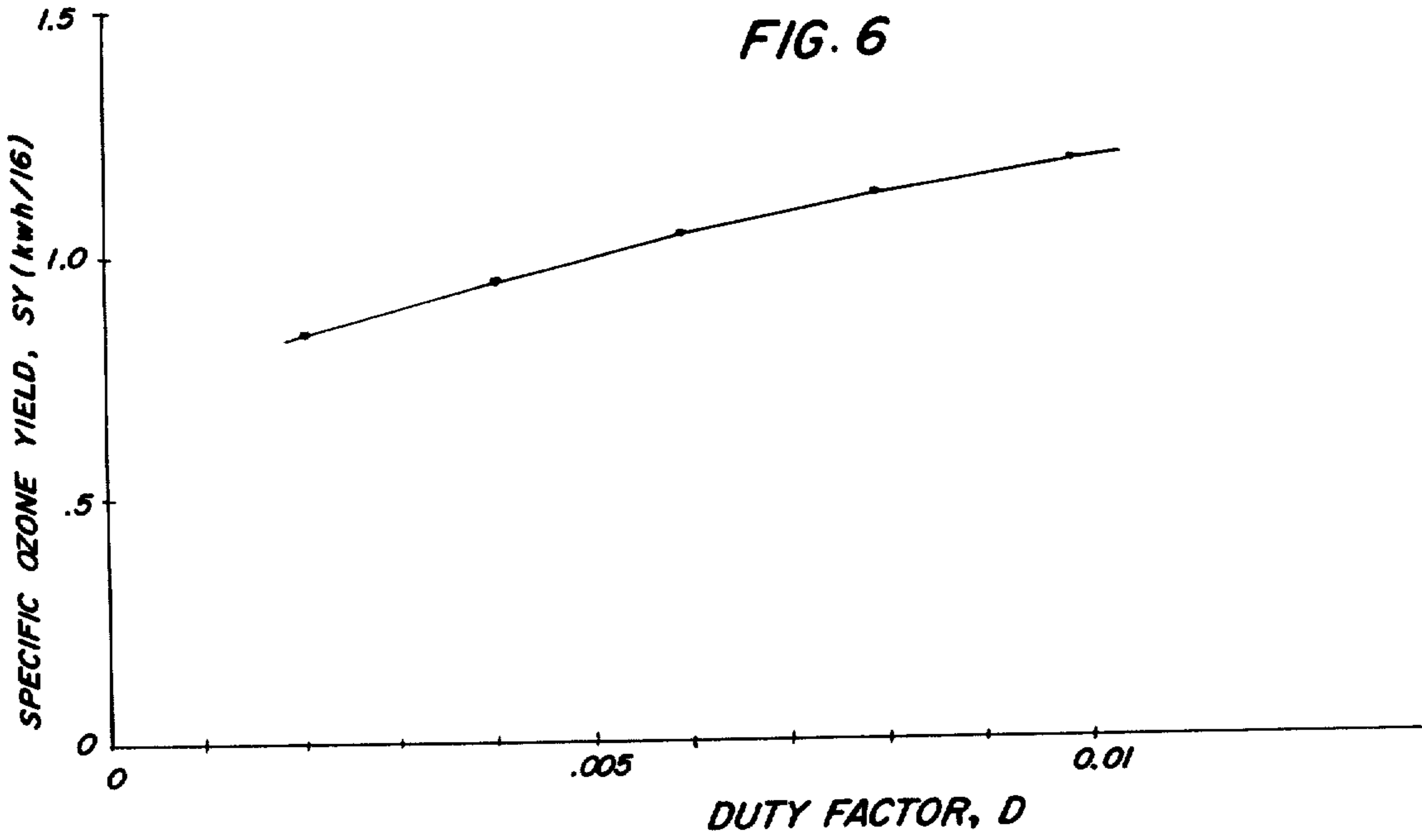
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18 Claims, 7 Drawing Figures





CORONA REACTION METHOD AND APPARATUS

Matter enclosed in heavy brackets [] appears in the original patent but forms no part of this reissue specification; matter printed in italics indicates the additions made by reissue.

The present invention relates to corona reaction systems, and more particularly to method and apparatus which may be used to increase the electrical efficiency of corona induced chemical reactions.

It is well known that many chemical reactions, such as the conversion of oxygen to ozone, may be effectively conducted in the presence of an electrical corona discharge. While it is found that high voltage corona is in many instances a convenient means by which to induce chemical reactions, corona discharge processes are extremely inefficient in terms of electrical energy required per unit of desired reaction product produced. For example, in the case of ozone produced from oxygen, the theoretical energy required to produce a Kg of ozone is 0.97 KWH per Kg while in practice it is found that about 6.6 KWH per Kg are needed with oxygen feed and 17.0 KWH/Kg with dry air feed.

Prior workers have attempted to decrease the amount of energy required to produce a corona reaction by varying many of the operational parameters of the corona generation system. For example, it is known that the use of pulsed high voltage energy having a pulse duration of 1 microsecond and a frequency as high as 1 Khz will more efficiently decompose carbon dioxide than a conventional 60 Hz AC waveform. However, it has also been shown that the use of an extremely high frequency corona power in the radiofrequency range, that is, 1-20 MHz, does not result in increased efficiency in the production of ozone.

While the prior art suggests that changes in efficiency may be obtained in corona induced chemical reactions by the manipulation of frequency and waveform, it has been found that these attempts have resulted in processes that show no improvement and are in fact impractical from the commercial standpoint. This is primarily due to the fact that the use of high frequencies and voltages result in the production of excess heat which is unmanageable when large scale operation and high unit capacities which require high power densities per unit of electrode surface are contemplated.

It is therefore an object of the present invention to provide an improved corona discharge system.

It is a further object to provide a method by which the overall electrical energy required to produce a corona discharge chemical reaction may be minimized.

It is still a further object to provide a means by which the amount of excess heat which results from a corona discharge reaction process is substantially decreased.

It is yet another object to provide a method by which the operation and equipment parameters involved in a corona discharge chemical reaction system may be optimized to produce maximum product and minimum waste heat.

It is still a further object to provide an improved corona generator system which will make feasible corona reaction processes which are conducted at high unit capacities and maximum electrical efficiencies.

It is still another object to provide an improved ozone generation system which is capable of efficiently pro-

ducing ozone in large quantities from an oxygen containing reactant gas, such as air, which contains considerable quantities of moisture and other impurities.

It is still a further object to provide economical, reliable, high voltage, high frequency narrow pulse power supplies which are capable of driving large size corona generators to produce commercial quantities of ozone.

It is still yet another object to provide a system by which ozone may be safely generated in concentrations of 10% by weight without undue decomposition of ozone.

These and still further objects of the present invention will become readily apparent to one skilled in the art from the following detailed description and drawings wherein:

FIG. 1 represents a cross-sectional view with parts broken away of a typical corona discharge cell which may be used in the practice of the present invention;

FIG. 2 is a graphic representation wherein applied, dielectric and gap voltages are plotted on the vertical axis versus time on the horizontal axis, and represents the preferred waveform of the electrical energy pulse used to generate corona discharge;

FIGS. 3 and 4 are circuit diagrams of preferred power supplies which may be used to obtain the desired corona discharge;

FIG. 5 is a schematic electrical circuit diagram which depicts a preferred series-parallel connection of a plurality of corona cells;

FIG. 6 is a plot in which specific ozone yield is plotted on the vertical scale versus duty factor on the horizontal scale, which may be used to select certain preferred operational parameters of the present system; and

FIG. 7 is a plot in which percent energy loss is plotted on the vertical scale versus duty factor on the horizontal scale, which may be used to select certain preferred operational parameters used in the practice of the present invention.

Broadly, my invention comprises a method for increasing the electrical efficiency of a corona discharge reaction system, and correspondingly substantially reducing the amount of waste heat produced thereby, wherein a narrow pulse high voltage waveform is applied along with a relatively low voltage bias potential.

More specifically, I have invented a corona reaction system wherein the corona is produced in a gas filled gap between opposing electrodes by a high voltage, narrow pulse electrical discharge in which the electrical pulse width is less than the gas ion transit time between the electrodes, and wherein a low voltage bias potential is maintained between the electrodes which is sufficient to substantially remove gas ions from the gap in the time interval between pulses.

In most conventional corona discharge systems the high voltage electrical potential is applied across the discharge gap in the form of relatively wide electrical pulses. Typically, the electrical energy waveform is a conventional sine wave, while in other instances pulse energy having a pulse duration of 1 to 200 microseconds (μ sec) have been utilized.

It is observed that these prior art systems are extremely inefficient and that 90 to 99% of the electrical energy is wasted in the form of excess heat. I have determined that the excess heat generated in these conventional systems may be attributed to the kinetic energy which is imparted to the charged gas molecules (ions) which are present in the corona discharge gap. The electrons which are responsible for the formation

of the desired reaction product are formed and accelerated to reaction potential during the initial part of the conventional electrical power pulse. The remainder of the pulse supplies kinetic energy to the charged gas molecules which result from the formation of electrons and which do not contribute to the formulation of reaction product. These ionized gas molecules dissipate their kinetic energy in non-productive collision which appears as waste heat in the reaction system.

In my process I have substantially reduced the amount of waste heat by modifying the applied electrical waveform in two ways.

1. The duration of the electrical power pulse (T_w) is selected to be substantially less than the gas ion transit time across the discharge gap (T_+).

2. A relatively low voltage bias potential (V_b) is maintained between the discharge electrodes to remove the majority of the charged gas molecules.

By selecting an extremely narrow energy pulse at the required sparking potential to produce ozone (V_s) it is found that the electrons which are formed are accelerated to an energy level required to do useful work. However, the energy pulse is not of sufficient duration to impart any significant waste energy to the charged gas molecules which are correspondingly formed.

It is recognized, however, that the gas ions which are present in the discharge gap must be removed before the next applied energy pulse, or a substantial portion of the energy pulse will be wasted in further acceleration of the non-productive gas ions. To achieve removal of gas ion "debris", I apply a relatively low voltage bias potential or "debris ion sweeping" potential during the interval between high energy electrical pulses. This low voltage bias potential is applied for a period which is substantially greater than the high energy pulse. However, since the energy applied to a charged particle in an electrical field is a function of only the particle charge and the field potential, the energy required to sweep the ion debris from the discharge gap is relatively minor.

The relationship which defines the preferred low voltage bias potential V_b is as follows:

$$T_r V_b = T_+ V_s$$

$$V_b = \frac{T_+}{T_r} V_s$$

wherein T_r represents the pulse repetition period, T_+ is the calculated gas ion transit time to sweep substantially all the gas ions from the gap at the sparking (corona discharge) potential V_s .

In order to determine the actual pulse width and frequency which is employed in the production of the high frequency narrow pulse energy it is necessary to select a pulse width which is substantially less than the pulse repetition period (which is the reciprocal of the [frequency] pulse repetition rate). In general, the actual pulse width T_w is selected so as to be a small percentage of the overall pulse repetition [rate] period T_r . In general it is found that in most applications the T_w will be from about 0.1 to 10 percent of T_r . Furthermore, it is noted that in order to produce the desired chemical reaction the pulse width should be of sufficient duration and magnitude to impart sufficient kinetic energy to electrons which produce the desired chemical reaction. Therefore, the desired narrow pulse width T_w will be somewhat greater than the pulse width required to

accelerate the electrons across the discharge gap, this time being referred to as T_e .

In order to implement my novel method and to optimize the design of the equipment and electrical circuits which are utilized herein, it is found that the operational and circuit parameters of the system may be conveniently selected in accordance with the following consideration of a circuit which employs a conventional heated cathode vacuum switch tube type power supply.

For most heated filament vacuum tube switch devices it is well known that the optimum electrical efficiencies are obtained when a duty cycle (D) is selected which is as high as possible. That is a relationship

$$D = \frac{T_w}{T_r}$$

is maximized commensurate with the energy losses which result in corona generation due to a pulse width T_w which is necessarily larger than the electron transit time T_e and substantially less than the gas ion transit time T_+ .

To optimize the parameters of duty cycle D and pulse width T_w , pulse repetition time T_r , which is the reciprocal of the pulse [frequency] repetition rate f , I have found the following relationship exists:

$$\alpha^2 = D \tag{1}$$

wherein:

$$\alpha = \frac{T_w}{T_+} \tag{2}$$

and T_+ is the gas ion transit time which may be readily calculated for any gas ion using the well known principles typically set forth in "Gaseous Conductors" by J. D. Cobine, Dover Publications, N.Y. (1958).

Equation (1) may be rewritten

$$D = \left(\frac{T_w}{T_+} \right)^2 \tag{3}$$

[For air] It is found by using the relationships provided by Cobine, that

$$\left[T_+ = \left(\frac{1}{K_p} \right) \frac{t_g}{40} \right] \quad T_+ = \left(\frac{1}{K_p} \right) \frac{t_g}{39.5 + \frac{1350}{P t_g}}$$

The product K_p is constant for any gas. For air

$$55 \quad K_p = [1064] \cdot 1034 \left(\frac{\text{cm/sec}}{\text{volt/cm}} \right) \text{ (mm Hg)} \tag{5}$$

A reasonable approximation of T_+ [is] for air at 1520 mm Hg.

$$T_+ = [1.83 \times 10^{-5} t_g] \frac{1}{1034} \times \frac{t_g}{39.5 + \frac{1350}{1520 t_g}}$$

Accordingly, to give an example of how the operational parameters may be optimized for a corona discharge system which is operating on air to produce

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ozone at a pressure (p) of 1520 m.m.Hg absolute with a discharge gap of 0.115 cm. the following calculations are made, using equation (6) above

$$[T^+ = 2.10 \mu\text{sec}] T^+ = 2.36 \mu\text{sec}$$

The sparking voltage V_s or gap potential required to produce corona is calculated from the following relationship, the basis for which appears in Cobine:

$$\begin{aligned} V_s &= 40 p \lg \\ &= 40 \times 1520 \times 0.115 = 6992 \text{ volts} \\ \hline V_s &= 39.5 p \lg + 1350 \\ &= 39.5 \times 1520 \times 0.115 + 1350 = 8250 \text{ volts} \end{aligned}$$

The pulse width T_w is determined using equation (2) above.

$$T_w = \alpha T^+$$

wherein α is less than 1. Furthermore, T_w is selected to be somewhat greater than the electron transit time T_e across the discharge gap which may be readily calculated by comparing the masses of the electron with the mass of the gas ion (oxygen) used in the calculation of the gas ion transit time T^+ . This calculation is as follows:

$$\begin{aligned} [T_e = \frac{T^+}{\sqrt{1840 \times 16}}] T_e &= \frac{T^+}{\sqrt{1840 \times 16}} \\ [= 0.012 \mu\text{sec}] &= 0.17 \mu\text{sec} \end{aligned}$$

It can be shown that the amount of power consumed in the production of ozone from oxygen, that is, specific ozone yield, S_y , which is due to pulse width losses and pulse bias losses is approximately,

$$\begin{aligned} [S_y = 0.6 (1 + \alpha) \left(1 + \frac{D}{\alpha} \right)] \\ S_y = 0.6 (1 + 4\alpha) \left(1 + 4 \frac{D}{\alpha} \right) \text{ KWH/lb. } O_3 \end{aligned}$$

and is a minimum when

$$\alpha = \sqrt{D}$$

Accordingly, for oxygen,

$$\begin{aligned} S_y (\text{min}) &= 0.6 (1 + 4 \sqrt{D})^2 \\ &= 0.6 (1 + 4\alpha)^2 \text{ KWH/lb. } O_3 \end{aligned}$$

and if S_y in KWH per pound of ozone produced is plotted against D , the duty factor, a curve such as is shown in FIG. 6 results. From FIG. 6 it is seen that D should be as low as possible to obtain the lowest ozone specific yield S_y in terms of KWH/lbs. O_3 produced as far as the pulse geometry is concerned. The expression "specific ozone yield" as used herein means the specific energy requirement for ozone production in KWH/lb. O_3 .

To select a specific value for the duty cycle D when using a typical heated cathode vacuum tube power supply circuit wherein the vacuum tube filament loss is of primary consideration, a plot such as is shown in FIG. 7 is prepared. In FIG. 7 the duty factor D is plotted as a solid line against the percent energy loss which is attributed to pulse geometry (which may be readily

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determined from FIG. 6). Also plotted are a series of broken lines in FIG. 7 which represent the percent energy loss due to filament heating in two typical vacuum tube switch devices rate at 100 and 150 KW respectively. It is seen from FIG. 7 that the preferred duty cycle D for the 150 KW tube is about 0.001 and it is approximately that point where the tube filament loss is equal to the pulse geometry, that is, process loss.

When the duty cycle D has been chosen to be 0.001 α is calculated as follows:

$$\begin{aligned} \alpha &= \sqrt{D} \\ &= \sqrt{0.001} \\ &= 0.0317 \end{aligned}$$

From α and T^+ the desired pulse width T_w and frequency f may be calculated as follows [:] for the air example:

$$\begin{aligned} [T_w = T^+ \\ = 0.0317 \times 2.09 = 0.066 \mu\text{sec}] \\ T_w = T^+ \\ = 0.0317 \times 2.36 = 0.066 \mu\text{sec} \end{aligned}$$

and

$$\begin{aligned} f = \frac{1}{T_r} = \frac{D}{T_w} \\ [= \frac{0.001}{0.066} 15100 \text{ Hz}] \\ = \frac{0.001}{.075 \times 10^{-6}} = 13300 \text{ Hz} \end{aligned}$$

To determine the preferred low voltage bias potential V_b which is used to remove positively charged gas molecules from the discharge gap between pulses, the following relationship is used:

$$\begin{aligned} V_b = \frac{T^+}{T_r} V_s \text{ and } T_r = \frac{1}{f} = \left[\frac{1}{15100} \right] \frac{1}{13300} \\ [V_b = \frac{2.09}{15100} \times 6975 \\ = 315 \text{ V.}] \\ V_b = 2.36 \times 10^{-6} \times 8250 = \frac{1}{13300} \\ = 260 \end{aligned}$$

In accordance with the above calculations, it is seen that the preferred operational parameters may be summarized as follows:

$$\begin{aligned} D &= 0.001 \\ V_s &= [6975 \text{ v}] 8250 \text{ v} \\ F &= [15100 \text{ Hz}] 13300 \text{ Hz} \\ V_b &= [315 \text{ v}] 260 \text{ v} \end{aligned}$$

A more clear understanding of the present invention may be obtained by reference to the drawings in which FIG. 1 is a cross-sectional view of a corona cell with parts broken away which may be used in the corona reactions systems contemplated herein. The corona reaction cell of FIG. 1 includes a gas tight housing 1 in which are placed two opposing electrodes 2 and 3. Between electrodes 2 and 3 are placed dielectric plates

4 and 5, the inner surfaces of which define a corona discharge gap having a thickness t_g , as indicated in FIG. 1. The dielectric plates 4 and 5 possess a thickness t_d , the actual dimension of which is enlarged considerably for purposes of clarity. Likewise, the dimension t_g is considerably expanded in the drawing for purposes of clarity. Typically, the dielectric thickness, t_d , may range as small as 0.1 mm and in certain instances may be as large as 24 to 50 mm. The discharge gap t_g may range from about 0.1 to 50 mm.

The electrode plates 2 and 3 are connected to an external power source through conductors 6 and 7 respectively, which enter the housing 1 through insulated bushings 7 and 8. While in the present drawing it is shown that the electrodes 2 and 3 are not in contact with the housing 1, it is found that in actual practice if the housing 1 is constructed of essentially electrically non-conducting material, the electrode plates may be in contact therewith. Furthermore, it is found in actual practice that either of the electrodes 2 or 3 may be grounded, and in the event the housing is constructed of conductive material, may support either of the electrodes.

As shown in FIG. 1, reactant gas enters the housing 1 through conduit 10 and the reactant product exits through conduit 11. The arrow 12 indicates direction of flow into the housing 1, while arrow 13 indicates the direction of the existing reaction products. Furthermore, in FIG. 1, 14 indicates the corona discharge zone which is defined between the electrodes 2 and 3 and the dielectric plates 4 and 5. While the present drawing shows the use of two dielectric plates, each of which is essentially in contact with the opposing electrodes 2 and 3, it should be understood that devices wherein the electrode plates are spaced from the electrodes, or wherein a single dielectric plate is located between, may also be utilized. In practice it is found that satisfactory results may be obtained when the dielectrics are constructed of materials having good dielectric qualities, that is dielectric strength of from about 100,000 to 1,000,000 volts per cm, such as glass. It is also found that extremely thin dielectric layers formed from fired on porcelain enamel coatings having a thickness of from about 0.1 to 0.5 mm produces particularly satisfactory results.

FIG. 2 shows in graphic idealized form the shape of the desired narrow electrical power pulses which are used in the practice of the present invention. It is noted that in FIG. 2 that 4 curves are plotted with voltage on the vertical axis and time on the horizontal axis. The curve E_1 (set forth as a solid line) represents the voltage of the preferred square wave pulse which is initially produced most efficiently by the high speed switch device of a suitable power supply. The curve E_2 (shown as a series of X's) represents the voltage of the initial power pulse E_1 after passing through a suitable inductance which has a value selected to minimize the power loss which normally results from the charging of a resistance-capacitance circuit. E_2 is the power pulse which is applied to the electrodes of the corona cell shown in FIG. 1. The curve V_g (dashed line) represents the voltage which appears across the discharge gap 14 of the corona cell and has a maximum value of V_s which is the sparking or ionization potential of the gap. V_g has a lower value of V_b which is the bias potential, a pulse width of T_w and a repetition period of T_r . The curve indicated as V_d (dotted line) is the value of the potential which appears across the dielectrics of the corona cell.

As will be discussed herein an understanding of the various preferred voltage wave forms which appear at various points in the corona cell will provide a basis by which one skilled in the art may design a circuit suitable for practice of the present invention.

While the preferred waveform shown in FIG. 2 indicates the pulses to be monopolar, it is to be understood that bipolar pulses in which each succeeding pulse is of alternating polarity may also be utilized. It is to be also understood that while FIG. 2 shows the bias potential to be greater than 0, that is, a + bias potential, it is found that V_b may also be utilized in the form of a negative bias potential, that is, a value less than 0. Furthermore, it is shown that V_b is essentially constant in the waveform shown in FIG. 2. It should be understood that V_b may be either increasing or decreasing between power pulses, that is, V_b does not necessarily have to remain constant, and in fact may alternate to some extent.

The waveform shown in FIG. 2 may be produced by a variety of well known high voltage power supplies which utilize high speed switch devices such as solid state transistors, silicon controlled rectifiers, varactors, vacuum tubes of both the heated filament and non-filament types, such as thyratrons and ignitrons, as well as high speed mechanical switches.

In FIGS. 4 and 5 two typical circuits are disclosed which use heated cathode vacuum switch tubes. It is to be understood, however, that the use of non-filament switch devices such as thyratrons and ignitrons have substantially no standby power losses and use thereof enables the operation of a more efficient process.

Reference to FIG. 3 reveals a corona generation system which includes a power supply of the typical vacuum tube amplifier type. In FIG. 3 a pulse generator 24 is connected to a DC power supply by means of electrical conduit 26. Pulse generator 24 is typically a commercial pulse generator unit such as is sold by Cober Electronics Inc., which is capable of producing pulse frequencies of a voltage up to 3000 volts at frequencies ranging from 1 Hz to 3 MHz. The pulse generator 24 typically comprises a trigger circuit in combination with suitable vacuum tube amplifier stages. The DC power supply 24 is also commercially available and typically produces from about 0 to +3000 volts DC at a power level of up to 10 kw.

Pulse generator 24 is used to control the output of vacuum switch tube 27. Vacuum switch tube 27 is a typical heated cathode tetrode device which is readily commercially available. Tube 24 includes a heated cathode 28 which is connected to a source of power by leads 29 and 30. Typically the heating power means, not shown, range from about 4 to 40 volts. The vacuum tube 27 is provided with a plate lead 32, a secondary grid lead 33 and a primary grid lead 34. The secondary grid 33 is connected to a source of bias potential which is confined within broken line 35. The bias potential supply comprises a diode 36 which is connected to a power transformer 37 which in turn is supplied through a variable resistance 38 by means of power leads 39 and 40 which are connected to a convenient 60 cycle AC source, not shown. The bias potential means 35 also includes a capacitance 41 which serves to even out the output thereof.

The switching grid 34 of the vacuum tube 27 is connected to the pulse generator 24. The plate lead 32 is connected to a DC power supply 45 through a conductor 46 which is connected to an inductance coil 47 in series with a resistance 48. The DC power supply 45 is

also connected through conductor 46 and conductor 54 in series with conductance coil 53 and conductor 52 to a corona cell 50. The coil 53 "spreads out" the square generated waveform such to minimize the energy loss normally associated with charging up a capacitor (cell capacitance in this case) through a charging resistor. In general, for low resistance circuits the value of the preferred inductance of the coil 53 is obtained by the readily recognized relationship

$$\left[L = \frac{t_w^2}{\pi C} \right] \quad L = \frac{t_w^2}{\pi^2 C}$$

A power supply 55 may be used to supply a bias potential to the plates of corona cell 50 through conductor 56 and inductance 57 which connect through inductance 53 and conductor 52. The capacitance 58 serves to isolate a DC bias voltage from the remainder of the power supply circuit. In the event the bias potential is supplied through the switch tube 27, the power supply 55 may be eliminated. The corona cell 50 is typically constructed in the manner shown in FIG. 1. The circuit through the corona cell 50 is completed by conductor 51 which is connected to the gate lead 32.

In operation the device of FIG. 3 requires that the output of DC power supply 45 be switched at a desired frequency which is produced by the pulse generator 24, which in turn is powered by the DC power supply 25. Typically, the DC power supply 45 is adjusted to operate about 5000 to 30,000 volts DC. The positive output side of the DC power supply 45 is applied to the corona cell as shown through the inductance 53 which typically has a value of 1.0 to 10.0 microhenry. The positive side of the power supply 45 also appears as being applied to the gate lead 32 through the inductance 47 which has a value of 0.1 to 1.0 henry and through the resistance 48 which has a value of 100 to 100,000 ohms. It is noted that switching of the output of vacuum tube 27 by means of the grid 34 which is connected to the pulse generator 24 produces a narrow high frequency pulse to appear across the plates of the corona cell 50. Typically, the pulse generator 25 is operated at a frequency of from 0.1 to 100 KHz. Also, for vacuum switch tubes of appropriate design the grid 33 of the power tube 27 is maintained at a suitable potential from about +50 to +500 volts so that a bias voltage of about 10 to 5000 volts may be maintained across the plates of the corona cell 50. Other methods of supplying a low pulse to pulse bias are possible, as shown in FIG. 4. In this manner the operational parameters of the corona cell in terms of frequency, bias voltage, sparking voltage and pulse width may be conveniently selected within the limits indicated by the present invention. The circuit of FIG. 3 is classically called a hard tube modulator.

Another suitable type of power supply is shown in FIG. 4, wherein a triode type vacuum switch type 60 includes a heated cathode 61, a plate 62 and a grid 63. This is a classical line pulser type modulator. The grid 63 is connected to a pulse generator 64 and a grounded conductor 65 which is also connected to the cathode 61. The cathode 61 is provided with heater leads 66 and 67 which are connected to a suitable source of power, not shown.

A high voltage DC power supply 70 is connected through its positive lead to the cathode lead 62 of the switch tube 60. The negative side of the DC power supply 70 is grounded. A DC bias voltage power supply

80 is negative grounded and the positive lead thereof is connected through an inductance 81 to conductor 82.

The DC power supplies 70 and 80 as well as the switch to circuit is connected to a conventional pulse forming network which is confined within the broken line 84 and includes selected inductances 85, 86 and 87 which are connected in series with each other through conductors 88, 89 and 90. Also included in the pulse forming network 85 are capacitances 91, 92 and 93 which are respectively connected to the conductors 88, 89 and 90 by means of conductors 94, 95 and 96. The pulse forming network 84 is connected to the DC power supply switch circuit by means of conductors 95 and 98. The output of the pulse forming network 84 is connected through to the primary side of a step-down transformer 100 by means of leads 101 and 102. The step-down transformer secondary is connected to a corona cell 105 by means of conductors 106 and 107. The step-down requirement is necessary at power levels greater than a few kilowatts. It is found to be impractical to build step-up pulse transformers with the required characteristics of band width, inductance, etc. at higher power levels. Step-down transformers are possible, however. The output of the bias power supply 80 is supplied to the corona cell 105 through the conductors 82 and 107.

In operation the system of FIG. 4 is adjusted so that the power supply 70 produces a voltage from about 10,000 to 100,000 volts DC which appears across the plate 62 of switch tube 60 and one side of pulse network 84. The DC power supply 80 is adjusted to provide a voltage of from about 10 to 10,000 volts DC which appears across the electrodes of corona cell 105. The pulse generator 64 is adjusted to provide a frequency output in the range of 0.1 to 200 KHz at a voltage of from about 100 to 10,000 v.

In the pulse forming network 84 the inductance 85, 86 and 87 are selected to have a value of from about 1.0 to 1000 microhenry, while the capacitances 91, 92 and 93 are selected to have a value of from about 10^{-6} to 1 microfarads. It is seen that by pulsing the pulse forming network from the switch tube circuit the output of the pulse forming network appears across the step-down transformer 100. Since the transformer 100 is of a step-down type, preferably the primary to secondary winding ratio will have a value of from about 1:2 to 1:100 and it is seen that the inductance characteristics of the power supply may be readily matched to those of the corona cell.

While for purpose of illustration the aforementioned systems of FIGS. 3 and 4 show the corona generator as a box and described as typically employing the corona or discharge cell of FIG. 1, it is found in many instances advantages to series several individual corona cells of the type shown in FIG. 1 to provide the voltage drop and impedance characteristics required by the high frequency power supplies of FIGS. 3 and 4. A particularly advantageous combination of corona cells is set forth in FIG. 5. It is seen in FIG. 5 that a multiplicity of corona cells 110 which are schematically shown as gas filled capacitance devices are series parallel connected by means of electric leads 111 and 112 which are connected through inductance coils 114 and 115 to power input lead 116. The opposite side of series connected corona cells 110 are connected to electrode input lead 117. Typically, leads 116 and 117 are connected to the power output leads 51 and 52 of the system shown in

FIG. 3 or the outlet leads 106 and 107 shown in FIG. 4, that is, the corona cell system of FIG. 5 may constitute the corona cell load 50 of the system of FIG. 3, and 105 the system of FIG. 4.

In the corona cell system of FIG. 5, the inductances 114 and 115 are selected so as to match the impedance of the series parallel connected corona cells with that of the output of the high frequency power supplies. Typically, inductance coils 114 and 115 will have a value of about from 1.0 to 100 microhenrys. While the system shown in FIG. 5 comprises two parallel connected corona cell banks, each bank consisting of three parallel connected lines of three series connected corona cells each, it is to be understood that the system may include as many parallel connected branches as desired. Furthermore, the number of series connected cells may be selected so as to provide the voltage drop required for the operation of any particular system.

It is found advantageous in the operation of many commercial systems to provide a means for series connecting the output of multiple corona cells in which the gas flow proceeds in series wise therethrough. Therefore, it is contemplated that in the system of FIG. 5 the reactant output of any given corona cell 110 may be series connected to the next and may include an intercooling heat exchange means which cools the reactant gas going from one corona cell to the other. It is also contemplated that in the system shown herein, intercooling of the output of one corona cell to the next may be conveniently eliminated due to the fact that the waste heat produced by the novel narrow pulse energy sources is minimized, therefore, intercooling may not be required except for every third, fourth and fifth series connected branch.

In the above I have outlined the basic aspects of the present invention. The following examples are given to illustrate the specific embodiments thereof.

EXAMPLE I

An ozone generating system was constructed which utilized the power supply circuit and arrangement generally set forth in FIG. 3. The pulse generator was a commercial Cober, Model 606, and the DC power supply, 25, was capable of producing 2000 volts DC at 1.0 amps. The DC power supply 45 had a capacity of 75,000 volts at 0.5 amps. The vacuum switch tube 27 was a 4 PR 100 A tetrode manufactured by Varian. The cathode heater leads 29 and 30 are connected to a source of 6.3 volts. The bias potential circuit 37 was regulated to produce a bias potential of +1600 volts. The inductance 47 had a value of 3 microhenry, whereas the resistance 48 was 2×10^6 ohms. The inductance 53 was 0.2 Henry.

The corona cell 50 comprised 3 cells connected in electrical series, each of which had an electrode surface area of 968 cm², a dielectric thickness of 0.44 mm, a gap dimension of 1.12 mm, wherein the dielectric comprised a fired-on porcelain enamel material. The corona cell was connected to a supply of air maintained at a pressure of 0.67 atmospheres (gauge) whereby a flow rate of 325 liters per minute through the corona cells was maintained. The pulse generator was operated at a frequency of 10,000 Hz with an output of +600 volts DC. The DC power supply 25 was operated at -500 volts DC, whereas the power supply 45 was operated at 30,000 volts. This produced a pulsed square power wave having the general shape shown in FIG. 2 wherein V_s was 6000, V_b was 200, T_w was 0.8 μ S and T_r was 100 μ S.

The device was operated under the above noted conditions wherein it was found that 450 grams of ozone per hour were produced the overall power consumption amounted to 2.2 kilowatt hours per kg of ozone

EXAMPLE II

The device described in Example I was operated under varying parameters, including feed gas, frequency, pulse width, bias potential and sparking voltage. A series of runs were made and the ozone produced and the power consumption required per [gram] kilogram of ozone produced were determined and summarized in the table set forth below.

Run No.	Gas	Gas Flow (liter/min)	Fray (H μ)	T _w (μ s)	V _s (Volts)	V _b (Volts)	Specific yield of KWH/kg O ₃
1	Dry O ₂	325	10,000	0.8	6000	500	2.2
2	Dry O ₂	325	5,000	0.8	6000	200	1.9
3	Dry O ₂	325	10,000	1.0	6000	600	2.4
4	Dry Air	325	10,000	0.8	6000	500	3.1
5	Wet Air (+10° C. DP)	325	10,000	0.8	6000	500	3.1

The above examples and description clearly indicate a method by which the overall electrical efficiency of a corona generation system may be improved. While it is noted that the above example is directed to the production of ozone, it is found that numerous other chemical reactions, yielding products such as N₂O₅, H₂O₂ and NH₃ may be conducted with equivalent increase in efficiencies.

I claim:

1. A process for conducting corona discharge reactions between spaced electrodes comprising the steps of:

- passing a reactant gas through a corona discharge gap formed between said spaced electrodes;
- periodically establishing a corona discharge by dissipating narrow pulse electrical power of high voltage in said gap with the width of each narrow pulse being a relatively small fraction of the pulse period as well as being less than substantially about the gas ion transit time across said gap and greater than the electron transit time; and
- maintaining a low voltage bias potential across said gap during the interval between said narrow pulses of a predetermined minimum magnitude sufficient to remove a substantial number of gas ions from said gap [being] before the termination of each pulse period.

2. The method of claim 1 wherein said reactant gas includes oxygen, and ozone is produced.

3. The method of claim 1 wherein said discharge gap has a width of from about 0.1 to 10.0 mm.

4. The method of claim 1 wherein each of said spaced electrodes is covered with a dielectric member having a thickness of 0.1 to 10.0 mm and a dielectric constant of 2.0 to 200 relative to vacuum; and a dielectric strength sufficient to withstand applied voltage.

5. The method of claim 1 wherein said gap is maintained at a pressure of -0.1 to 3.0 atmospheres (gauge).

6. The method of claim 2 wherein said gas is maintained at a temperature below about 100° C.

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7. The method of claim 1 wherein said power has a frequency of 0.1 to 200 KHz.

8. The method of claim 1 wherein said high voltage lies in a range between 2.0 to 200 k volts.

9. The method of claim 1 wherein the narrow pulses have a duration from about 1 to 50 percent of the singly charged gas ion transmit time.

10. The method of claim 1 wherein said low voltage bias potential is selected from the group consisting of positive, negative and/or alternating potentials having a magnitude of from about 0.1 to 50 percent of said high voltage.

11. The method of claim 1 wherein said pulses are supplied from a square wave power supply through an inductance.

12. The method of claim 11 where said pulses are of the same polarity.

13. The method of claim 11 wherein said pulses are of alternating polarity.

14. The method of claim 1 wherein said pulses are produced by vacuum switch tube power supply in which the filament energy losses are substantially equal

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to the energy consumed in removing the gas ions with said low voltage bias potential.

15. The method of claim 1 wherein the reactant gas is moist air.

5 16. A process for producing ozone by corona discharge which comprises:

a. passing oxygen through a corona gap having a width of 0.1 to 10 mm;

b. producing a corona discharge by applying electrical power at a peak voltage of about 2.0 to 200 k volts and a frequency of 0.1 to 200 kHz, said power being applied as high voltage pulses where said pulses have a duration of about 0.1 to 10 percent of the pulse repetition period; and

c. maintaining a low bias potential across said gap between said high voltage pulses of about 0.1 to 50 percent of said peak voltage.

17. The method of claim 16, wherein said bias potential is applied for a period of about 10 to 100 percent of said pulse repetition period.

20 18. The method of claim 17 wherein said bias potential is substantially equal to the product of the oxygen ion transit time and the peak voltage divided by the pulse repetition time.

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