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Ozone Production in a High Frequency Dielectric Barrier Discharge Generator

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Abstract

Factors that affect the performance of an expanded-mesh dielectric barrier discharge ozone cell were investigated. A gas feed pf 94% O_2, 4% Ar and 1% N_2 was used. An improvement in the productivity (g ozone/kWh) of about 20 % was achieved by doubling the gas flow rate through the cell. Decreasing the cell operating frequency (in the range 72 kHz to 19 kHz) increased the productivity of the ozone generator at constant power. The ozone production increased approximately in proportion to the input power; however productivity did not vary significantly with power above a minimum level. As the cell voltage was increased the dependence of productivity on power or frequency was reduced. Changing the feed gas temperature between -5°C and +42°C had no effect on productivity. Finer meshes drew more power than coarser ones for a given voltage. Using a thinner mesh for the centre electrode increased productivity. The best results were obtained with a 6x3x1.86 mm titanium mesh giving a productivity of 110g ozone/kWhr at 30-60W, 1500-1900V and 23 KHz.

Key Words

Ozone; Ozone Generation; Dielectric Barrier Discharge; High Frequency; Expanded Mesh Dielectric Barrier Discharge;

Introduction

Surface discharge is the most common method of producing ozone particularly by dielectric barrier discharge (1-9), which has the highest energy efficiency and hence the lowest running costs. This makes it the preferred option for large-scale ozone generation. A dielectric barrier discharge ozone generator consists of two electrodes separated by a solid dielectric and a discharge gap. A feed gas flows through the discharge gap between the two electrodes, where the electrical discharge occurs. This requires high voltages producing small discharge currents and only occurs at gas pressures near atmospheric. Some of the oxygen is dissociated into atomic oxygen, which then forms ozone. As only displacement currents can travel through the dielectric, the voltage must vary with time.

For dielectric barrier discharge cell power is dependent on the electrode spacing. If local parts of the electrodes are closer together a more intense discharge develops in this region and the higher local power draw leads to local heating. The higher local temperature will cause higher ozone destruction in this region, reducing the overall efficiency of the ozonator (2;10;11). Thus to maximize efficiency, the discharge should be homogeneous.

Most commercially available dielectric barrier discharge ozone generators operate at frequencies up to 5 kHz (6;7;12-15). A high frequency ozone generator has been patented that can operate at frequencies of up to 100 kHz (16) and is characterized in this study. The advantage of increasing the frequency of discharge is that this allows the power input to the ozone generator to be raised. The same amount of ozone can then be generated from a smaller cell. In addition, the increase in frequency allows a reduction in the size of the power supply (16-18).

The high frequency Novozone cell uses an expanded metal mesh as one electrode (Figure 1). Typically the mesh electrode has a sinusoidal voltage of 32 kHz (between 20 kHz and 100 kHz) and up to 3000V rms applied between it and the two ground electrodes. The discharge occurs on both sides of the mesh, making the unit compact and maximizing the proportion of the gas volume in the cell that is subject to electrical discharge. It is also safer as both outside electrodes are grounded (16). Thousands of small randomly distributed micro-discharges occur in the gas between the dielectric and the electrode (8). The discharges occur over larger gaps as the cell voltage is increased. The function of the dielectric is to spread the discharge evenly over the electrodes and to separate the electrodes (16). The mesh electrode touches the dielectric so the ozone generator's discharge is a mixture of surface and silent discharge (19). This complex discharge may offer improved performance over traditional ozone generators when high humidity feed gases are used (17).

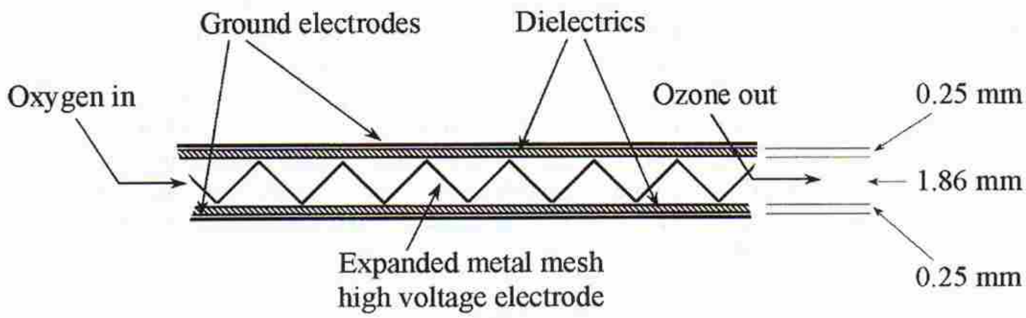

Figure 1. The Ozone Cell Arrangement Studied (for a 10x6 mm mesh - not to scale)

In this study the factors that affect the performance of the cell are investigated. The objective was to understand how to maximize the ozone output per cell and the energy efficiency of the cell. A number of variables were investigated and are described in this paper. These were the effect of power, gas pressure, gas flow rate, frequency and power, cell voltage and power, inlet gas temperature, and mesh size on the production of ozone.

Only one gas composition was used in these tests. However the effect of impurities such as nitrogen, argon, carbon dioxide, SF_6 and water on ozone production has been reported in other systems. Small amounts of nitrogen improve the ozone production at low power inputs compared to using pure oxygen. (2;3;14;20-22). Addition of SF_6 to air improves ozone production (9) while water vapor has a detrimental effect on the production of ozone (23;24).

Experimental Methods

A Novozone cell was used in the tests and modified for some experiments as described below. The feed gas for ozone generation was produced by pressure swing adsorption at a concentration of between 93.5 and 94% oxygen (the remainder of the gas is argon 5 % and nitrogen 1.0-1.5 %). The gas was stored in a tank from which it is delivered for the experiments. The feed pressure, normally 150kPa, was measured by a VDO pressure transducer, the temperature by a thermocouple in the feed line, and the flow rate, normally 6 liters per minute (lpm), with a Porter rotameter (variable area) flow meter. Mass flow rate was measured and reported in terms of standard liters per minute (i.e. liters at 1 atm) rather than liters of gas at the pressure used. After passing through the ozone cell the gas stream was split with 1 lpm going to the ozone monitor, the rest being exhausted to atmosphere, or to vacuum.

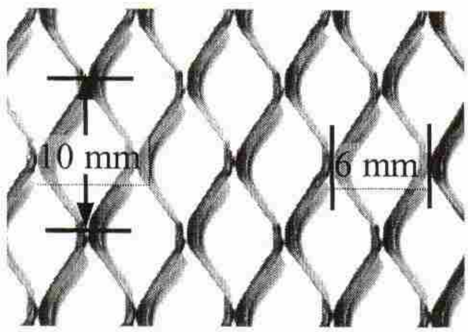

Figure 2. A Section of 10 x 6 mm Mesh

The ozone concentration was measured with a PCI HC 400 ozone monitor, which measures the UV adsorption of the product gas by comparing the UV adsorption of the ozonated gas at 254 nm with that of reference gas from the surge tank.

The performance of the cell is defined in terms of "productivity". By this we mean d(ozone production)/ d(energy) and use units of g ozone/kWhr.

The temperature of the feed gas was varied between -5°C and +42°C to assess its effect on ozone production by passing feed gas through a copper coil before entering the cell. The coil was placed inside a vessel and media of various temperatures added. Hot water, cold water and ice, and a mixture of solid carbon dioxide and automotive anti freeze gave the range of inlet temperatures. The gas temperatures at the inlet and outlet of the cell were measured by thermocouples inserted into the gas connection fittings.

A variety of mesh sizes for the centre electrode were used to determine the effect of changing the mesh size on ozone production and productivity. Expanded metal meshes are characterized by their diamond size and mesh thickness. Diamond size is the distance between knuckles along and across each the diamond shaped hole. Mesh thickness is largest at the knuckles. All the meshes were tested with the longest diamond dimension perpendicular to the gas flow. Normally the ozone generator has an expanded titanium mesh, similar to the one shown in Figure 2. Titanium mesh was not available in any other sizes, so aluminum meshes were used. All experiments, except those investigating the effect of mesh size, used the 10x6 mm titanium mesh with a thickness of 1.86 mm. To look at the effect of mesh size aluminum meshes were tested with diamond sizes 6 x 3, 10 x 6, 12.5 x 6 and 20 x 6 mm, and thickness' of 1.34, 1.86, 2.0 and 2.2 mm respectively, since titanium mesh was not readily available in a range of sizes. The performance of the aluminum mesh was compared with the titanium mesh at one size (10 x 6 mm). The electrodes had an area of 160 x 60 mm. Changing the mesh size affected cell capacitance and the gas volume in the cell and hence the flow velocities and gas residence time for a particular flow rate. To compensate for the capacitance change, capacitors were added in parallel with the cell. Aluminum channel was used to make the two outer electrodes. The dielectric used was a 0.25 mm thick mica-silicone composite.

The power supply was a resonant circuit in which the operating frequency was altered by adding inductors and capacitors in parallel with the secondary winding of the power supply transformer. The cell voltage, operating frequency and power were measured using a Tektronix TDS 210 oscilloscope. One channel of the oscilloscope was connected to a 50:1 voltage divider, the other channel was connected across a capacitor in series with the cell. These waveforms were transferred to computer and the cell power calculated using the Lissajous figure method (8).

Results and Discussion

The Effect of Flow on the Productivity of Ozone Production

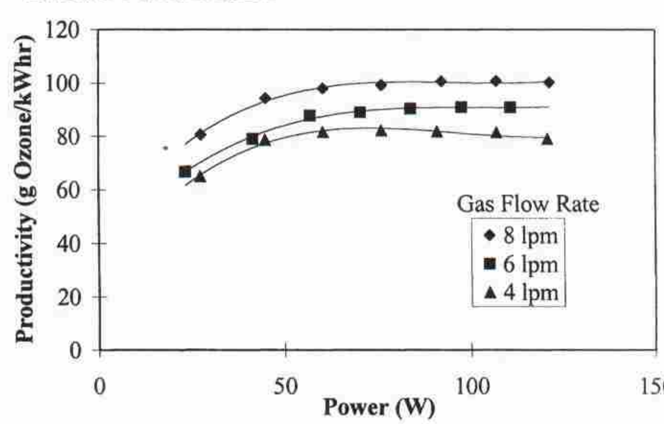

Figure 3. The effect of gas flow rate on ozone productivity (150 kPa and 32 kHz)

Ozone production was measured at gas flow rates of 4 to 8 standard liters per minute. As the flow rate was increased an improvement in the productivity (ozone production per unit energy) of about 20 % is achieved (Figure 3). It is believed this is due to the decrease in gas residence time so that the ozone that is produced has less opportunity to further react with the electric discharge and be destroyed, in accordance with accepted theory (2). The increased gas velocity is also likely to result in a lower localized temperature near the mesh and decrease the rate of thermal ozone destruction. There is a slight decline in productivity at high power input for the 4 lpm flow however this was not observed at higher flow rates (Figure 3).

The Effects of Power and Pressure on Ozone Production

Ozone production was measured at pressures of 150-300 kPa and cell power of 20-120 W (power densities of 2.1-12.5 kW m^{-2}). The flow rate of gas through the cell was held steady at four standard liters per minute and therefore the residence time in the cell varied with pressure. As the power input was raised the ozone production increased almost linearly as would be expected (Figure 4).

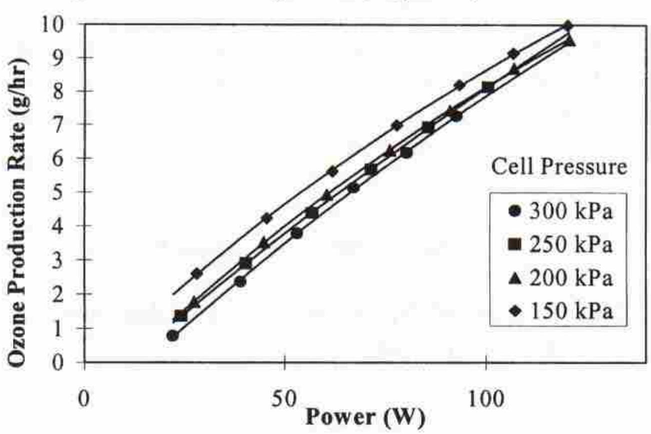

Figure 4. The effect of power on ozone production (150 kPa, 4 lpm, 32 kHz)

However, at high power input, the rate of increase of ozone production with power begins to decreases slightly. It is believed that this is due to two factors: High power leads to high cell temperatures. The resultant rise in cell temperature causes the rate of thermal destruction of ozone to increase. High power also means higher ozone concentrations and an increase in the rate of collision of ozone with energetic electrons. The amount of ozone lost by collision is expected to be proportional to both the discharge current and the ozone concentration so that this higher cell power would result in more ozone lost by this process.

At the lowest pressure (150 kPa) the ozone production was highest. Ozone production was found to fall with increasing pressure at all power inputs (Figure 5). A doubling of the pressure results in around a 20% drop in the production, which accords with the change in the residence time of the gas in the cell. As the pressure is increased the residence time of the gas in the cell increases. This will result in the ozone being exposed to the discharge for a longer period of time, which will cause more of the ozone to be destroyed.

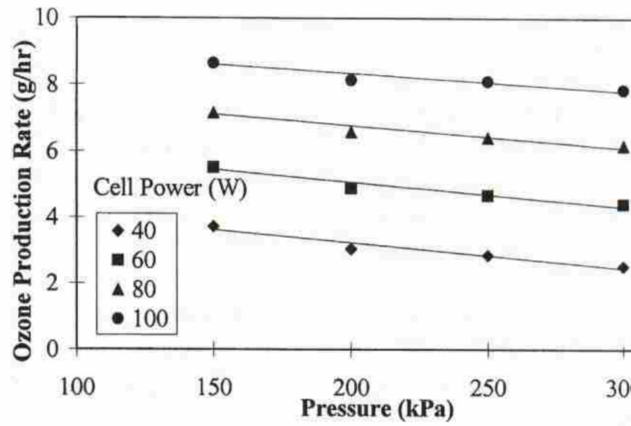

Figure 5. The effect of pressure on ozone production rate (4 lpm, 32 kHz)

Over the range of frequencies studied, the productivity is approximately constant with input power, above a minimum power level of 40 - 60 W. This suggests that the power input to the cell could be further increased without significantly reducing the productivity. This would increase the ozone production of a unit without affecting its physical size.

The Effect of Frequency and Cell Voltage on Ozone Production and Productivity

The ozone production and productivity were measured at frequencies between 19 and 72 kHz at a flow rate of 6 standard lpm and a pressure of 150 kPa (Figure 6). The operating frequency, with our particular power supply arrangement, is set by the resonant frequency of the cell and power transformer secondary winding. As the power input to the cell is increased this frequency rises slightly, so the results are not exactly at the frequencies given.

In the frequency range studied, the productivity of ozone generation falls as the operating frequency is increased, with the highest productivity being encountered at the lower frequencies. This suggests that to operate at the highest productivity the frequency should be in the lower range. An earlier study of an ozone generator operated at frequencies between 300 Hz and 3 kHz found, at constant cell power, frequency had no effect on production or productivity (25). We might therefore suppose that there is an optimum frequency for maximum productivity somewhere at or below 19 kHz.

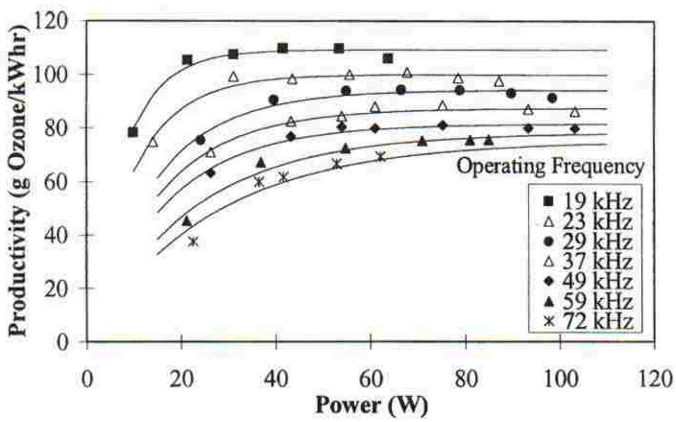

Figure 6. The effect of frequency and power on productivity - empirical curves fitted (6 lpm, 150 kPa).

There are disadvantages in operating at these frequencies as low as 19 kHz, however, as the noise from the ozone generator becomes audible and irritating. In addition, the voltage must be higher in order to achieve he same power input, which would require more insulation and make the manufacture of the power supply more expensive.

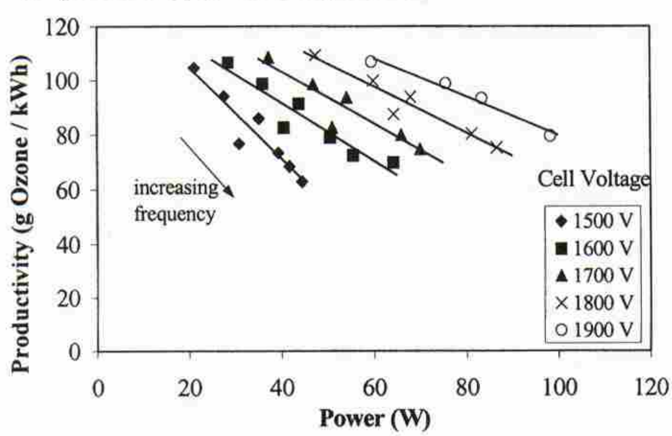

Figure 7. The effect of cell voltage and power on productivity (6 lpm, 150 kPa and at variable frequency since the frequency must be increased to increase the power at constant voltage)

The operating frequency also influences the cell power. To keep the power constant as the frequency is raised the cell voltage must be reduced. By interpolating between the experimental results a plot of the influence of cell voltage has been prepared (Figure 7). When extrapolated, these plots of have a common intercept at zero power of about 138 g/kWhr which represents the maximum potential productivity of the cell in the absence of secondary degradation reactions of ozone with the electrical discharge.

The Effect of Inlet Gas Temperature on the Production of Ozone

The ozone productivity was measured at inlet temperatures between -5 and 42°C with a flow rate of six standard liters per minute and pressure of 150 kPa. The productivity results have been interpolated to prepare a plot of ozone productivity against inlet temperature for various power inputs (Figure 8). With a change in inlet gas temperature from -5°C to 42 there was no significant change in productivity at all power inputs.

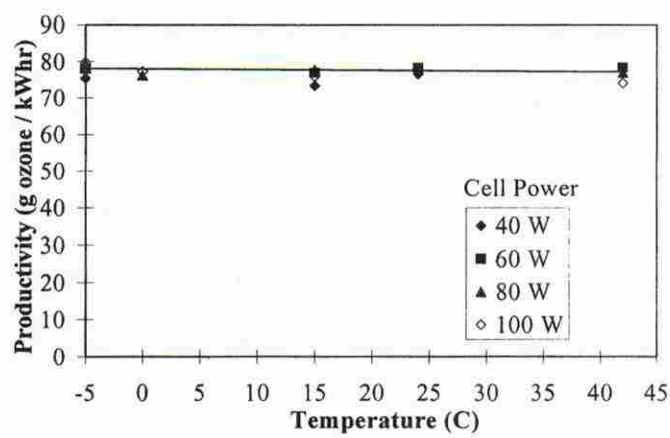

Figure 8. The Effect of Inlet Temperature on Ozone Productivity (6 lpm, 150 kPa, 49 kHz)

This is perhaps not surprising since the energy removed from the feed gas when it is cooled to -5°C from room temperature is approximately 3.8 mW which is insignificant compared to the power consumption of the ozone generator of 20-80,000 mW. Thus the cold feed gas is immediately heated as it enters the ozone generator, negating the effect of cooling, so the gas inlet temperature has no effect on the outlet temperature. Maintaining a low cell temperature is a very important factor in maximizing ozone productivity but this can be best achieved by concentrating on heat removal through the walls of the cell.

The Effect of Different Mesh Sizes on Ozone Production

Four aluminum meshes were tested, at a pressure of 150 kPa and flow of six liters per minute, to assess the effect of mesh size on ozone production and productivity (Figure 9). Since all the other experiments were carried out with titanium mesh we also compared the aluminum mesh to the titanium mesh for the 10x6 size.

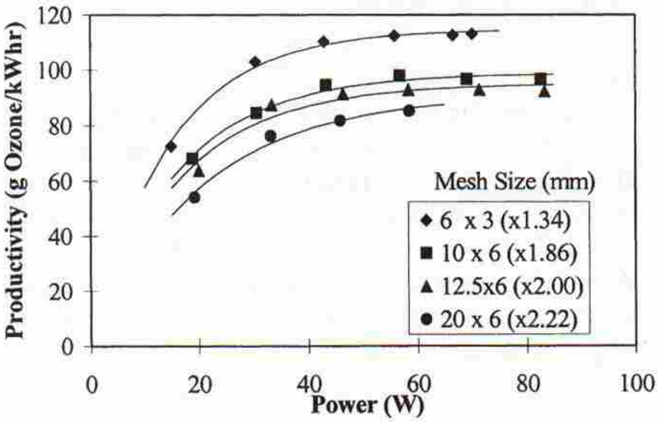

Figure 9, The Effect of Mesh Size on Ozone Production (6 lpm, 150 kPa, 23 kHz)

The productivity when using the 10x6 aluminum mesh is similar to that obtained for the titanium mesh. Productivity is not affected by changing between these two materials. Aluminum is not normally used in the ozone generator, however, because of its lower resistance to corrosion by water vapor.

The smallest mesh (6x3 mm) has the highest productivity (Figure 9). Using a small mesh, with a small gap, and therefore small gas volume in the cell, reduces the gas residence time for equivalent flow rates.

This may be due to the change in the electric discharge. The thinner mesh has a larger amount of electrode area close to the dielectrics, so the electrical discharge is spread over a larger proportion of the mesh. This gives a larger real volume in which electrical discharge takes place. Not only does this spread the power over a larger area to lower localised temperatures but it also reduces the amount of gas that can bypass the electric discharge.

The closer spacing of the electrodes affects the power drawn (Figure 10). Power draw increases with increasing mesh size at constant voltage. The 6x3 mm mesh draws the highest power because it has more electrode area close to dielectrics (hence having a small air gap to breakdown).

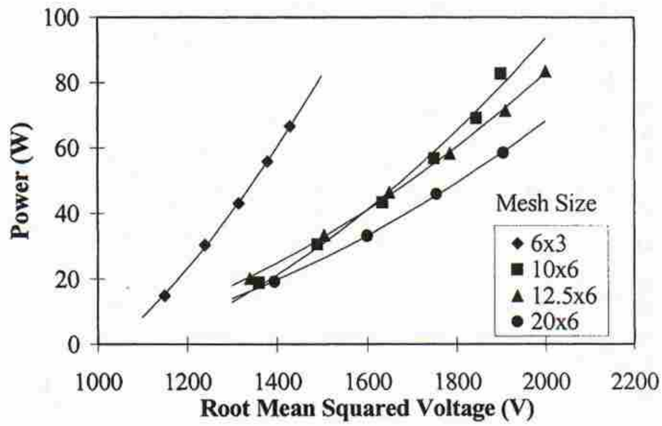

Figure 10. Power Drawn by Various Meshes (6 lpm, 150 kPa, frequency variable)

The desirable characteristics of the fine meshes could be used to advantage to decrease the cell voltage or operating frequency. Getting the same power into the cell at a lower voltage would allow a reduction in transformer insulation. Operating at lower frequencies, but similar voltages would increase productivity without requiring changes to the transformer design. The higher flow velocities for the same flow rates also improve the productivity. The gas flow has been modeled in this system and will be the subject of a subsequent paper.

Conclusions

The factors that affect the performance of an expanded mesh dielectric barrier discharge ozone cell were investigated in order to understand how to maximise the ozone output per cell and the energy efficiency of the cell. These factors were the effect of power, gas pressure, gas flow rate, frequency and power, cell voltage and power, inlet gas temperature, and mesh size on the production of ozone.

An improvement in the productivity (g ozone/kWh) of about 20 % was achieved by doubling the flow rate. The decreased gas residence time may mean that the ozone that is produced has less opportunity to further react with the electric discharge to be destroyed. Also the localized gas temperature is lower at higher flow rates also reducing ozone decomposition.

The ozone production increases approximately in proportion to the input power. Over the range of frequencies studied, the productivity is approximately constant with input power above a minimum power level of 40 - 60 W. When extrapolated to zero power a productivity of 138 g/kWhr is calculated which represents the maximum potential productivity of the cell in the absence of secondary degradation reactions of ozone. The power input to the cell could be further increased without significantly reducing the productivity. This would increase the ozone production of a unit without affecting its physical size.

Ozone production was found to fall with increasing pressure at constant standard gas flow rates (in the range 150 kPa - 300 kPa) at all power inputs. However, this could be attributed to the change in residence time of the gas.

Decreasing the cell operating frequency (in the range 72 kHz to 19 kHz) was found to increase the productivity of the ozone generator at constant power. To achieve the same power input at a lower frequency, however, the cell voltage must be higher. This would make the manufacture of the power supply more expensive. In addition, if the frequency is lowered below about 20 kHz the noise from the unit becomes audible.

As the cell voltage is increased the dependence of productivity on power (and therefore frequency) is reduced. To get the highest productivity the ozone generator should be operated in the lower frequency range and at a high voltage

Changing the feed gas temperature between -5°C and +42°C had no effect on productivity. This is because the energy removed from the gas is insignificant compared to the cell power. To improve productivity by cooling, heat should be removed from the cell, not the feed gas.

It was found that using thinner mesh for the centre electrode increases productivity. This may be due to the change in the electric discharge with electrical discharge spread over a larger proportion of the mesh. The mesh size was also found to influence the power consumption of the ozone generator. At the same voltage finer meshes drew more power than larger ones. This effect could be used to reduce the cell voltage, decrease transformer insulation, or lower the operating frequency without changing the transformer design

The best results were obtained with a 6 x 3 x 1.86 mm titanium mesh giving a productivity of 110g ozone/kWhr at 30-60W, 1500-1900V and 23 KHz.

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