

Ozone generation at high pressures

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The present invention relates to a method for producing ozone at elevated pressure having the features of the preamble of Claim 1.

High pressures at the outlet of the ozone generator of up to 12 bar are needed in some areas of application of ozone. This includes, for example, the
10 bleaching of paper in the paper industry. Ozone generators with ozone capacities of several kg/h at working pressures of around 1-2 bar are generally used to bleach paper.

The unit "bar" will be used as a technical unit of pressure in this description and claims. This is to be understood as excess pressure over atmospheric
15 pressure. The numerical value of the absolute pressure which is to be distinguished from this is 1013 mbar greater.

An ozone generator of this type is for example known from application document WO/2013/136663. A tubular high voltage electrode is surrounded by a tubular dielectric and a tubular counter electrode. The electrodes are
20 arranged in a concentric manner relative to one another. The dielectric is on the inside of the counter electrode. A gas containing oxygen flows through the gap between the dielectric and the high voltage electrode. Silent discharges develop between the dielectric and the high voltage electrode during operation. The distance between the dielectric and the high voltage electrode
25 is called the gap width or stroke width. The ozone generator can be operated at a gas pressure of between 1.2 and 3.2 bar with a gap width of 0.5 mm. If the gap width is decreased by 0.2 mm, the pressure range is between 2 and 5.3 bar. The voltage necessary for ignition increases while the gap width remains the same as the gas pressure increases. This effect can be

compensated for by decreasing the gap width. For production-related reasons, the gap in this type of ozone generators cannot be arbitrarily decreased to ensure operation at high pressures. Furthermore, only power supplies of up to around 20 kV can be used due to insulation problems. Conventional ozone
5 generators are therefore limited to a pressure range as a result of the maximum voltage and the minimum gap width.

The profitability of ozone generation also plays an important role. This type of ozone generators no longer reach ozone concentrations that are as high as the pressure increases while the efficiency of the ozone generation decreases as
10 the pressure falls. Economic ozone production and the generation of high concentrations of ozone is therefore not possible at high pressures. Conventional ozone generators have an optimal operating pressure depending on the ozone concentration ("Effects of discharge gap width and gas pressure on ozone generation characteristics of an Air-Feed-Ozone generator", J.
15 Kitayama et al., Proceedings 13th Ozone World Congress, Kyoto). Any deviations from this optimal operating pressure result in considerable reductions in efficiency ("Ozone production in a high frequency dielectric barrier discharge generator", R.G. Haverkamp et al., Ozone Science & Engineering Vol. 24, pp 321-328). Higher operating pressures are therefore
20 uneconomical for conventional ozone generators.

Water ring compressors are used to compress the ozone to the necessary pressure. However, these compressors are expensive to buy and consume high quantities of energy.

The object of the present invention is to provide a method for the production
25 of ozone at elevated pressure in which high ozone concentrations and high ozone capacities can be achieved effectively.

This object is achieved by a method with the features of claim 1.

According to this a method is provided for the production of ozone at elevated pressure with a capacity of at least 1 kg ozone/h by means of an ozone
30 generator having a high voltage electrode and at least one counter electrode, wherein the high voltage electrode and the at least one counter electrode

delimit a gap in which at least one dielectric is arranged and through which a gas containing oxygen and having a gas pressure of p_{gas} flows, and wherein the high voltage electrode and the at least one counter electrode with a connection for an electric power supply to generate discharges are provided in
5 at least one discharge gap, wherein the power supply provides a voltage in a range from 1 kV to 50 kV and wherein stroke widths d_i of the discharge are distributed between a minimum stroke width d_{min} and a maximum stroke width d_{max} , wherein the gas pressure p_{gas} of the gas containing oxygen at the outlet of the ozone generator is at least 3 bar. This method can be used to produce
10 ozone at elevated pressure with a high ozone concentration and high ozone capacity in an economic manner without using water ring compressors.

The power supply preferably provides a voltage in the range of between 10 kV and 30 kV, preferably between 15 kV and 20 kV. Power suppliers of this magnitude can be used in industry and are accordingly cost-efficient.
15 Furthermore, an ozone concentration of at least 10% by weight is preferably achieved.

In an embodiment the gas pressure p_{gas} of the gas containing oxygen is at least 6 bar and in an advantageous embodiment the gas pressure p_{gas} is between 10 bar and 12 bar.
20 The gas containing oxygen is preferably at least 80% O_2 and more preferably at least 98% O_2 . This means high ozone concentrations can be produced with good energy efficiency.

The ozone is preferably produced with a production rate of at least 20 kg of ozone/h and in particular 100 kg of ozone/h. At this capacity, an ozone
25 generation system of the type mentioned above is particularly effective.

It is advantageous if the gas containing oxygen flows into the gap at a feed-in rate in the range from 500 kg/h O_2 to 800 kg/h O_2 .

In an advantageous embodiment the minimum stroke width is zero mm. The stroke width distribution is preferably continuously and periodically or
30 stochastically distributed.

The ozone generator preferably has its own counter electrode and the dielectric is arranged in the system on the counter electrode. This arrangement is a classic one-gap system.

The distribution of stroke widths is preferably formed by a fabric made of wire.

- 5 The term "fabric" (woven or non-woven fabric) is known from textiles technology. A textile fabric can refer to any planar structure made from textile raw materials using a textile technology. Accordingly, in the context of the present patent application a fabric is understood to mean any flat, curved or convex planar structure manufactured using a textile technology. These
10 include, inter alia, non-woven fabrics, such as tissue, knitted fabrics, meshes and nets, and fibre composite fabrics such as non-wovens and cotton wool.

It is advantageous if the fabric made of wire fills the gap.

- The high voltage electrode is preferably formed at least in part from the fabric. It is also possible to provide for the fabric forming the full high voltage
15 electrode.

A preferred embodiment of the invention is described below in more detail with reference to the drawings. In the drawings:

- Fig. 1: shows a perspective view of an electrode arrangement from the prior art and
20 Fig. 2: shows a basic progression of the energy consumption of a conventionally operated ozone generator and an ozone generator operated with the method according to the invention by gas pressure.

- Figure 1 shows an electrode arrangement as known from DE 10 2011 008
25 947 A1. Ozone generators of this type are used in an ozone generator in groups. The ozone generators are thereby arranged parallel to one another between two tube plates in the manner of a tube bundle heat exchanger and connected in an electrically parallel manner. The ozone generator shown has a tubular outer electrode 1, an also tubular dielectric 2 and an
30 internal rod 3, wherein shortened versions of the individual components

are shown stretched out from one another in an axial direction. The arrangement is rotationally symmetrical. The outer electrode 1, the dielectric 2 and the rod 3 are oriented concentrically to one another. There is a wire mesh 4 between the outer electrode 1 and the dielectric 2 that fills the gap. Accordingly, a wire mesh 5 is provided between the dielectric 2 and the rod 3 which also fills the gap there. The outer electrode 1 is designed in the form of a stainless steel tube. The waste heat generated during the ozone production is cooled by means of cooling water which is passed along the outer side of the outer electrode between the bases of the tubes. The dielectric 2 is a glass tube. The wire meshes 4 and 5 are preferably manufactured as what are known as circular hollow strings made of a stainless steel wire mesh. The rod 3 arranged in the centre of the electrode arrangement is an insulator, for example made of glass of another material compatible with oxygen and ozone. The rod 3 can be designed to be massive. During operation, pressure is exerted on the electrode arrangement by means of a gas containing oxygen having a gas pressure of 1 to 2 bar which flows through the wire meshes 4 and 5 in the direction of the arrow 6. An electric power supply 7 is schematically indicated, which power supply is in contact with the outer electrode 1 on the one side and with the mesh 5 on the other. The operating voltage supplied by the power supply 7 a silent electrical discharge in the space between the electrodes 1, 5 and the dielectric 2 which generates ozone from the oxygen flowing through the meshes 4 and 5 in the direction of the arrow 6.

In the structure shown the inner electrode is formed solely from the mesh 5 while the rod 3 exercises a supporting function as an insulator which ensures the even filling of the inside of the dielectric 2 with the wire mesh 5. This shape of the electrode results in an overlapping of volume and surface charge.

Figure 2 shows curves of the specific energy consumption of ozone generators.

With reference to the method according to the invention,

$$U(t) = U_0 * \sin(2\pi * f * t)$$
even at comparatively high gas pressures and voltage amplitudes (in the range indicated), there are still surface points on the high voltage electrodes from which silent discharges originate. It is surprising in

this context that the specific energy consumption of the ozone generator measured in kWh/kg of ozone is significantly lower as a result of this operation according to the invention at high pressures compared to conventional ozone generators 11. The above-mentioned characteristic optimal operating pressure for ozone generators described above depending on the specific energy consumption is in a narrow range about the minimum of a parabolic distribution (see curve 11). In contrast to this, an ozone generator with a profiles high voltage electrode has a significantly flatter distribution (see curve 10). The specific energy consumption is still comparatively low at high pressures, for example of more than 3 bar, or even higher pressures. Since the minimum distribution is flatter, the operating pressure can be in a wide range, for example in a range from 1 to 12 bar with almost the same level of efficiency in terms of ozone generation.

The advantage of this method is further that no compromise has to be made between gas pressure and capacity and/or ozone concentration.

According to the invention the ozone generator is operated at capacities of one to several hundred kg of ozone/h. In one embodiment the ozone concentration is at least 12% by weight ozone.

The method according to the invention is therefore suitable for ozone production in the paper industry as high ozone concentrations and high ozone capacities can be produced effectively at high pressures.

The high voltage electrode is an electrically conductive material, preferably stainless steel with a profiled surface. The high voltage electrode can be a wire mesh or a knitted fabric, a fabric or even a wire winding or a granulate placed on a surface. Fibrous fabrics such as files or felts are also suitable as structures that are placed on the electrode by means of mechanical processing or by coating. The profiling is stochastically or periodically distributed and is distributed in both a longitudinal and a peripheral direction of the electrode.

It is also conceivable, however, to profile the dielectric instead of the electrode, with the same effect being able to be achieved.

The method according to the invention is not limited to tubular electrode arrangements. It can be used in both tubular and flat ozone generators. Use in single-gap and multi-gap systems is provided for. The electrically conductive material of the electrode can be introduced into the discharge gap with or
5 without carrier material.

The method of ozone generation according to the invention can of course also be used more generally in plasma generators.

The ozone generator can be adapted to customer-specific requirements by means of the method according to the invention. This is economical due to the
10 increasing energy prices, and is also ecologically advantageous. Ozone generation at the high pressures at the outlet of the ozone generator of up to 12 bar are technically and economically possible. The water ring compressors that are usually required are therefore not necessary.

C l a i m s

1. Method for the production of ozone at elevated pressure with a capacity
5 of at least 1 kg ozone/h by means of an ozone generator having a high
voltage electrode (5) and at least one counter electrode (1), wherein the
high voltage electrode (5) and the at least one counter electrode (1)
delimit a gap in which at least one dielectric is arranged and through
which a gas containing oxygen and having a gas pressure of p_{gas} flows,
10 and wherein the high voltage electrode (5) and the at least one counter
electrode (1) with a connection for an electric power supply (7, 8) to
generate discharges are provided in at least one discharge gap, wherein
the power supply (7, 8) provides a voltage in a range from 1 kV to 50 kV
and wherein stroke widths d_i (9) of the discharge are distributed between
15 a minimum stroke width d_{min} and a maximum stroke width d_{max} ,
characterised in that the gas pressure p_{gas} of the gas containing
oxygen at the outlet of the ozone generator is at least 3 bar.
2. Method according to Claim 1, **characterised in that** the power supply
(7, 8) provides a high voltage in the range from 10 kV to 30 kV.
- 20 3. Method according to Claim 1, **characterised in that** the power supply
(7, 8) provides a high voltage in the range from 15 kV to 20 kV.
4. Method according to any one of the preceding claims, **characterised in
that** the method is designed such that the ozone produced has an ozone
concentration of at least 10% by weight.
- 25 5. Method according to any one of the preceding claims, **characterised in
that** the gas pressure p_{gas} of the gas containing oxygen is at least 6 bar.
6. Method according to any one of the preceding claims, **characterised in
that** the gas pressure p_{gas} of the gas containing oxygen is between 10
bar and 12 bar.

7. Method according to any one of the preceding claims, **characterised in that** the gas containing oxygen is at least 80% O₂.
8. Method according to any one of the preceding claims, **characterised in that** the gas containing oxygen is at least 98% O₂.
- 5 9. Method according to any one of the preceding claims, **characterised in that** the method is designed such that ozone is produced at a production rate of at least 20 kg of ozone/h.
- 10 10. Method according to any one of the preceding claims, **characterised in that** the method is designed such that ozone is produced at a production rate of at least 100 kg of ozone/h.
11. Method according to any one of the preceding claims, **characterised in that** the gas containing oxygen flows into the gap at a feed-in rate in the range from 500 kg/h O₂ to 800 kg/h O₂.
- 15 12. Method according to any one of the preceding claims, **characterised in that** the minimum stroke width d_{min} (9) is zero.
13. Method according to any one of the preceding claims, **characterised in that** the ozone generator has a single counter electrode (1) and the dielectric (2) is arranged on the counter electrode (1).
- 20 14. Method according to any one of the preceding claims, **characterised in that** the distribution of the stroke widths (9) is formed by a fabric (5) made of wire.
15. Method according to Claim 14, **characterised in that** the fabric (5) made of wire fills the gap.