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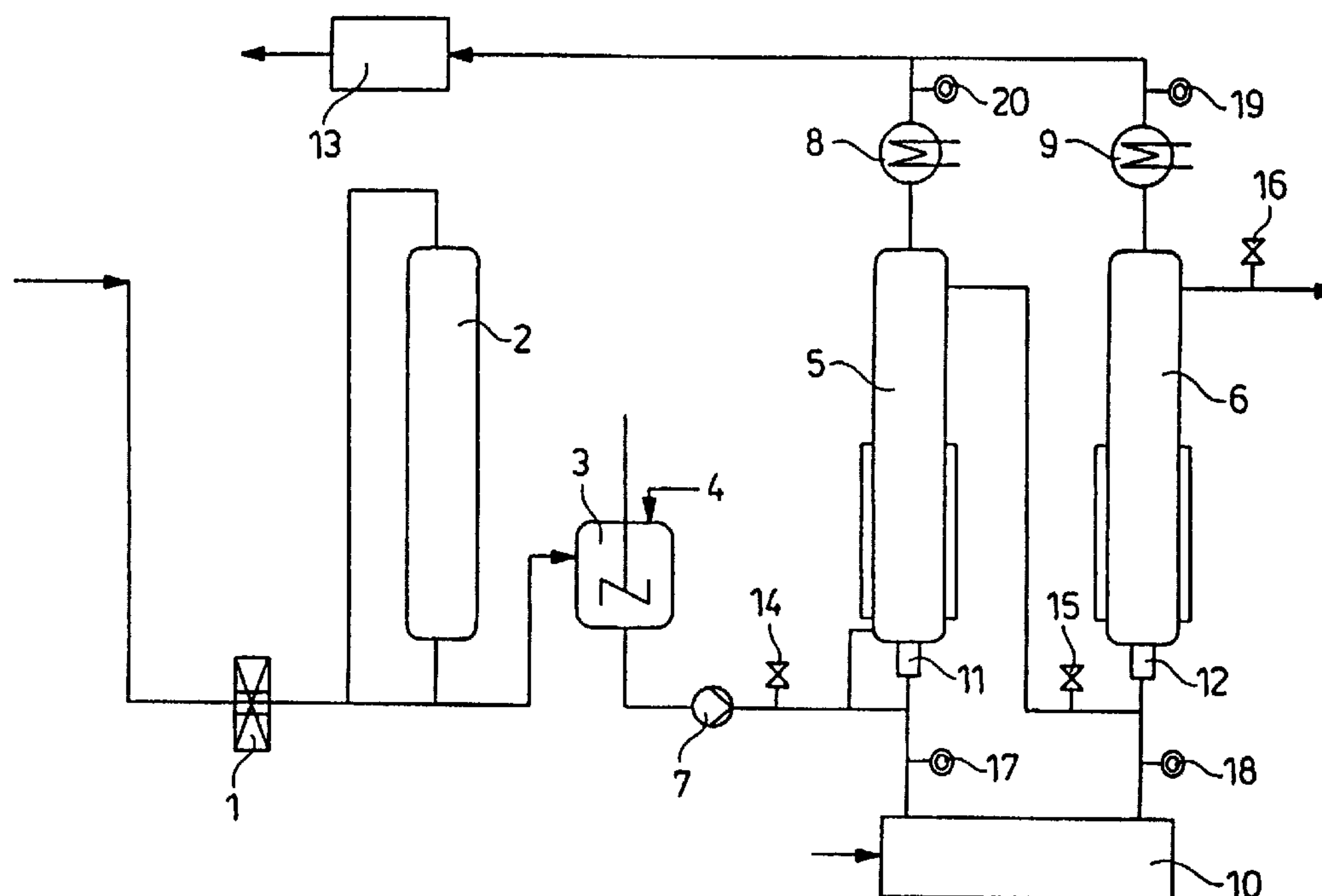
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(54) Titre : PROCEDE DE DEGRADATION DE COMPOSES ORGANIQUES DANS L'EAU

(54) Title: METHOD OF DECOMPOSING ORGANIC COMPOUNDS IN WATER



(57) Abrégé/Abstract:

The invention relates to a method of decomposing organic compounds in water which has a TOC content of more than 2 ppm and a content of carbonic acid and carbonates dissolved therein, by treating the mixture with ozone. The invention further relates to a method of producing chlorine by subjecting sodium chloride to an electrolysis. The inventive method is characterized in that the sodium chloride is used in said electrolysis in the form of an aqueous solution that is obtained by treating water with ozone, said water having a TOC content of more than 2 ppm and a sodium chloride content of 2 to 20 wt.-% and additional carbonic acid and carbonates dissolved therein.



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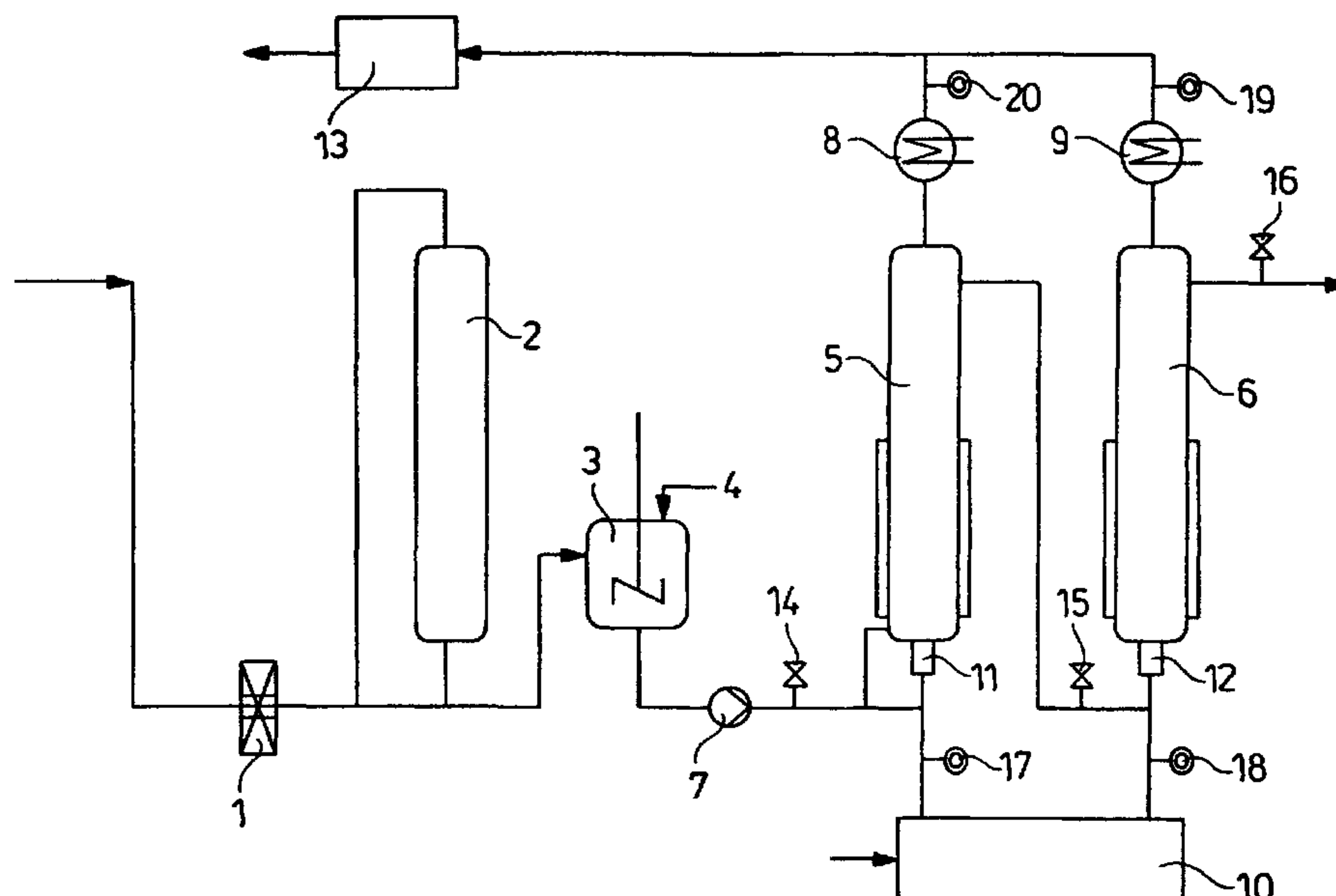
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[Fortsetzung auf der nächsten Seite]

(54) Title: METHOD OF DECOMPOSING ORGANIC COMPOUNDS IN WATER

(54) Bezeichnung: VERFAHREN ZUM ABBAU ORGANISCHER VERBINDUNGEN IN WASSER



(57) **Abstract:** The invention relates to a method of decomposing organic compounds in water which has a TOC content of more than 2 ppm and a content of carbonic acid and carbonates dissolved therein, by treating the mixture with ozone. The invention further relates to a method of producing chlorine by subjecting sodium chloride to an electrolysis. The inventive method is characterized in that the sodium chloride is used in said electrolysis in the form of an aqueous solution that is obtained by treating water with ozone, said water having a TOC content of more than 2 ppm and a sodium chloride content of 2 to 20 wt.-% and additional carbonic acid and carbonates dissolved therein.

[Fortsetzung auf der nächsten Seite]

Process for the decomposition of organic compounds in water

The present invention relates to a process for the decomposition of organic compounds in water which has a TOC of more than 2 ppm and contains in addition dissolved carbonic acid or carbonates, by treatment with ozone, and a process for the production of chlorine by electrolysis of common salt, characterised in that the common salt is added to the electrolysis process in the form of an aqueous solution obtained by treatment with ozone of water which has a TOC of more than 2 ppm and a common salt content of 2 to 20 wt.% and contains in addition dissolved carbonic acid or carbonates.

Processes for the decomposition of organic compounds in water by treating the water with ozone are known.

WO 9708101 describes the treatment of industrial waste water with ozone in the presence of a catalyst. EP-A 634465 describes the purification of industrial waste water with ozone in a two-step process, wherein aromatic compounds in particular are decomposed. EP-A 378994 describes the decomposition of aromatic impurities in industrial waste water by ozone at increased pressure and elevated temperature.

As described, for example, by E. Gilbert in Water Res., Volume 21 (10), pages 1273-1278, the use of ozone in industrially contaminated waste water generally serves to convert impurities consisting of compounds which are not, or only inadequately, bacterially degradable into components which are bacterially degradable. This means that even after the treatment with ozone, the water contains organic compounds which, in subsequent steps such as a biological waste-water treatment, then have to be decomposed into inorganic compounds such as, for example, carbon dioxide and water.

This is confirmed by Takahashi in Ozone Science and Engineering, Volume 12, 1990, pages 1 to 18. Takahashi reports that the treatment with ozone of water which contains

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phenol results in the decomposition of the phenol, but that organic decomposition products such as, for example, oxalic acid, glyoxal and glyoxalic acid are formed which are not further decomposed by ozone.

5 If water contains organic compounds, this indicates a certain carbon content, which is present in the form of organic compounds. The carbon content which is present in the form of organic compounds is referred to as TOC (abbreviation of "Total Organic Carbon").

10 Water having a TOC of more than 2 ppm and containing in addition dissolved carbonic acid or carbonates is known, for example, in polycarbonate production.

To produce polycarbonates by the so-called phase interface process, dihydroxydiaryl-
alkanes in the form of their water-soluble alkali metal salts are reacted with phosgene
15 in heterogeneous phase in the presence of inorganic bases such as, for example, sodium hydroxide solution and with an organic solvent in which the polycarbonate product is readily soluble. During the reaction, the aqueous phase is distributed in the organic phase. After the synthesis of polycarbonate by the phase interface process, the polycarbonate is separated in the form of its solution in the organic solvent used in the
20 synthesis, for example, methylene chloride. The remaining aqueous phase is advantageously freed from volatile organic impurities, such as remains of the organic solvent used in the synthesis, for example, methylene chloride; this can be effected by distillation, for instance. There then remains waste water having a high content of dissolved carbonates (for example, 0.3 to 1.5 wt.%) and a high content of dissolved
25 common salt (for example, 4 to 12 wt.%). Furthermore, the waste water is contaminated with organic compounds such as, for example, phenols (for example, unsubstituted phenol or alkylphenols or arylphenols or bisphenols such as, for example, bisphenol A) or amines (for example, triethylamine or ethylpiperidine). The carbonates are formed in the course of this, for example, by the hydrolysis of phosgene
30 as a secondary reaction in the polycarbonate production process.

The common salt dissolved in the waste water from polycarbonate production by the phase interface process is a valuable raw material. A possible way of utilising the common salt in the waste water from polycarbonate production by the phase interface process has not as yet been described.

5

A conceivable way of utilising the load of common salt in the waste water from polycarbonate production by the phase interface process is to use the common salt for the production of chlorine and sodium hydroxide solution by electrolysis. However, this possible method has hitherto failed because of the other constituents, in particular the organic constituents, of the waste water from polycarbonate production by the phase interface process. In particular, the especially advantageous membrane process for chlor-alkali electrolysis requires pure aqueous solutions of common salt as starting material.

10

If the common salt is added to the electrolysis process in the form of an aqueous solution, this aqueous solution has to contain a low concentration of organic impurities; the TOC of the common salt solution must preferably be less than 1 ppm. Even when the TOC of the common salt solution is less than 1 ppm, the common salt solution may still to a small extent contain organic impurities which interfere with the electrolysis process, for example, by decreasing the useful lives of important components of the plant such as, for example, the membranes in the case of the membrane process. All this applies in particular to the membrane process for the electrolysis of common salt, where the useful life of the membranes is an important factor which determines the economic efficiency.

20

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Of course, other waste waters, which do not arise from polycarbonate production and are characterised in that they contain dissolved carbonates or carbonic acid, are also known.

30

It is now known that carbonates or carbonic acid dissolved in the water hinder the removal of the organic compounds from the water by treatment with ozone because

carbonate acts as a free-radical scavenger and thereby hinders the decomposition of organic compounds via radical intermediate steps. This is reported by Hoigne and Bader in Wat. Res., Volume 10, 1976, page 377 ff. and by Gurol and Watistas in Wat. Res., Volume 21, 1987, pages 895 to 900.

5

For water which contains organic compounds and in addition dissolved carbonates or carbonic acid, there consequently arises the situation that the treatment with ozone in the alkaline pH range is hindered by the carbonate ions. In the acidic pH range, however, the treatment with ozone, according to the reports from prior art, results not in the complete decomposition of the organic compounds, but in the formation and persistence of compounds such as, for example, oxalic acid. In both cases, therefore, a complete decomposition of the organic compounds and hence of the TOC to inorganic decomposition products such as, for example, carbon dioxide and water, by treatment with ozone is impossible according to prior art.

15

Accordingly, the object of the present invention is to provide a process for decreasing the content of organic compounds in water which contains dissolved carbonic acid or carbonates.

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If the water contains dissolved common salt, the object of the present invention is also to provide a process for decreasing the content of organic compounds in the water containing the dissolved carbonic acid or carbonates to such a small amount that the common salt dissolved in the water can be used for the production of chlorine by electrolytic processes.

25

The object according to the invention is achieved by a process for the treatment, with ozone, of water which has a TOC of more than 2 ppm and contains at least 0.01 wt.% of dissolved carbonic acid or carbonates, characterised in that the treatment is carried out at a temperature of 10°C to 130°C and at an absolute pressure of 0.5 to 3 bar and that the water which is supplied to the process has a pH value of 2 to 11 and that the treatment takes place over a period of from 1 minute up to 10 hours.

30

It has also been found that the water treated according to the invention, if it contains dissolved common salt, ~~can~~ be used for the production of chlorine and sodium hydroxide solution by electrolysis of the common salt. In particular, the chlorine can
5 be produced by the well-known membrane process. Here the process is not interfered with by impurities which may possibly still be present in very low concentration; in particular, the useful life of the membranes is not decreased in comparison with the use of common salt solutions which have been obtained by dissolving common salt in completely pure water.

10

The process according to the invention for the treatment of water with ozone is particularly economic, is not expensive technologically and is non-polluting. A high excess pressure is not required. No catalyst is required. No UV irradiation is required. No additional chemicals such as, for example, hydrogen peroxide are required. Of
15 course these measures can however, additionally be carried out.

The process according to the invention allows the TOC in the water to be decreased to below 1 ppm.

20 The water treated with ozone according to the invention is moreover so pure that it can be introduced directly into surface water without the necessity of a further purification. An economic and environmentally favourable possible method of working up and disposing of water which has a TOC of more than 2 ppm and in addition dissolved carbonic acid or carbonates is thereby provided.

25

According to the invention, the TOC of the water prior to the treatment with ozone is more than 2 ppm, preferably more than 5 ppm, particularly preferably more than 10 ppm.

30

In the invention, the TOC is determined in accordance with DIN 38 409 - H 3, using a TOC 500 apparatus from Shimadzu, from a measurement of the content of inorganic

carbon (TIC) and of the content of inorganic or organic carbon (TC) in the water sample. A constant flow of highly pure air, free from carbon dioxide, is used as carrier gas. For the TC measurement, a given quantity of the sample to be analysed is injected into the TC combustion tubes and is burnt there at 680°C on a TC catalyst. The carbon dioxide formed, after cooling and drying, is detected in an infrared analyser. For the TIC measurement, the sample is acidified with phosphoric acid and the carbon dioxide formed is expelled from the sample and detected as above. The TOC is then calculated from the measured TIC and TC values as follows:

$$\text{TOC} = \text{TC} - \text{TIC}.$$

10

According to the invention, the content of carbonic acid or carbonates in the water is at least 0.1 wt.%, calculated as carbonate (CO_3^{2-}). It is preferably at least 0.3 wt.%, particularly preferably at least 1.0 wt.%.

15 The treatment with ozone according to the invention takes place at a temperature of 10°C to 130°C, preferably at 20°C to 100°C, particularly preferably at 60°C to 90°C.

The treatment with ozone according to the invention takes place at an absolute pressure of 0.5 to 3 bar, preferably at 1 to 2 bar, particularly preferably at 1.2 to 1.8 bar.

20

The water supplied to the process according to the invention for the treatment with ozone has a pH value of 2 to 11; preferably it has a pH value of 3 to 11, particularly preferably it has a pH value of 5 to 9 and most particularly preferably it has a pH value of 5.5 to 7. The pH value is measured at 20°C.

25

A particularly preferred embodiment of the invention is provided by ensuring that the pH of the water supplied to the process for the treatment with ozone has a value which is less than 7 and is such that, after the treatment of the water with ozone, the pH value is more than 7.5. This shift in the pH value from the acidic to the basic range in the course of the treatment with ozone results in a particularly effective decomposition of

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the TOC in the water. To achieve this shift in the pH value from the acidic to the basic range, the pH value of the water supplied to the process for the treatment with ozone has to be adjusted according to the carbonate content of the water and the possible content of other substances which give rise to a change in the pH value in the course of the treatment of the water with ozone.

The treatment with ozone according to the invention is carried out over a period of from 1 minute up to 10 hours, preferably from 6 minutes to 2 hours, particularly preferably from 10 minutes to 60 minutes.

If the water which is subjected to the treatment with ozone according to the invention contains dissolved common salt, a preferred embodiment of the present invention is a process whereby, after the ozone treatment of the water, the latter is then passed to the electrolysis step for the production of chlorine. Here the electrolysis is preferably carried out by the membrane process. The water which is subjected to the treatment with ozone according to the invention contains, for example, 2 to 20 wt.% common salt. It preferably contains 4 to 12 wt.% common salt.

The production of chlorine by electrolysis of common salt is described, for example, in Ullmann's Encyclopedia of Industrial Chemistry, Volume A 6, 5th Edition, 1986, pages 401 to 477. In particular, the especially advantageous membrane process is also described here on pages 437 to 450.

A precondition for the utilisation of the common salt in the waste water from polycarbonate production by electrolysis is the decomposition of the organic matter contained in the waste water to values below approximately 1 ppm. This applies in particular when the especially advantageous membrane process is to be used for the chlor-alkali electrolysis.

The water which is subjected to the treatment with ozone according to the invention is preferably the waste water from the production of polycarbonate by the phase interface

process, and particularly preferably waste water from the production of bisphenol A polycarbonate by the phase interface process.

5 The temperature of the water supplied to the ozone treatment process can be adjusted to the required value, for example, by means of a heat exchanger.

10 The pH value of the water supplied to the ozone treatment process can be adjusted to the required value, for example, by adding an acid such as, for example, hydrochloric acid, or a lye such as, for example, sodium hydroxide solution.

15 To achieve an optimal reaction, the water and the ozone produced in the ozone generator should be mixed together as intensively as possible. For this, in principle any form of gas distribution is possible, for example, the use of ultrasound, glass frits or conventional injectors.

20 The oxidation of the waste water with ozone is preferably carried out in a continuously operated process, with preferably several reaction columns being connected one behind the other. To achieve as quantitative a decomposition of the TOC as possible, preferably a substream of the freshly produced ozone is passed directly to the columns following the first column.

25 A preferred embodiment of the invention is provided by using at least two reaction columns, in particular three reaction columns, for the treatment with ozone and in the course of this varying the ozone distribution between a distribution of 1:1 or 1:1:1 (volumetric flow rate of first column: volumetric flow rate of second column) in each column up to 5:1 or 5:1:1. The ozone distribution in the reaction columns is preferably 80% in the first column(s) and 20% in the last column.

30 Peak loadings of the TOC possibly occurring in the water can be adsorbed, for example, by an adsorption column connected upstream of the plant for the treatment with ozone and under normal operating conditions can be passed in a well-regulated

manner to the ozone treatment process. The adsorption column is preferably only operated in cases where loading peaks occur.

5 The adsorption column is preferably constructed as follows. The jacket of the adsorption column is cooled by water. An operating temperature of 15°C is thereby established. The adsorption column is filled with adsorbers that adsorb phenol and bisphenol well and, depending on the pH, also desorb phenol and bisphenol again, such as, by way of example and preferably, a microporous, non-functionalised, hydrophilic, hypercrosslinked copolymer based on styrene and divinylbenzene, or
10 active charcoal as adsorber.

The process according to the invention renders possible, for example, the treatment of the waste water from the production of polycarbonate by the phase interface process. The purified waste water can then be used for the exploitation of the common salt
15 contained in it for the production of chlorine by electrolysis. Here it is advantageous to increase the concentration of the salt in the water which can, for example, amount to 4 to 12 wt.%, by addition of solid common salt to a concentration of 20 to 30 wt.%, preferably 25 wt.%, before the water is passed to the electrolysis. The chlorine produced during the electrolysis and the sodium hydroxide solution can again be
20 passed to the process for the production of polycarbonate by the phase interface process, after the chlorine has been reacted with carbon monoxide to form phosgene and the sodium hydroxide solution has been used, for example, for the preparation of bisphenolate solutions.

25 The organic compounds which constitute the TOC contamination of the water may be any organic compounds. These can include both aliphatic and aromatic compounds. The compounds may contain all kinds of hetero atoms. In the case of the treatment of waste water from polycarbonate production, the organic compounds consist substantially of phenols (for example, unsubstituted phenol or alkylphenols or
30 arylphenols or bisphenols such as, for example, bisphenol A) and amines (for example, triethylamine, ethylpiperidine).

The ozone used in the process according to the invention is produced, for example, from air or from oxygen by known processes. In the course of this, mixtures of ozone in air or in oxygen are formed which contain, for example, 40 to 150 g ozone per cubic
5 metre of gas and which can be used as such. Higher concentrations of ozone can be produced by special concentration processes (adsorption processes or desorption processes).

Unused ozone which is present, for example, in the waste gas from the plant for the
10 treatment of water with ozone, can be thermally or catalytically decomposed in a residual-ozone annihilator.

Other preferred embodiments of the present invention are provided by carrying out in addition a treatment with hydrogen peroxide prior to, during or after the treatment with
15 ozone. The treatment with hydrogen peroxide is preferably carried out at the same time as the treatment with ozone.

Other preferred embodiments of the present invention are provided by carrying out in addition a treatment with UV radiation prior to, during or after the treatment with
20 ozone. The treatment with UV radiation is preferably carried out at the same time as the treatment with ozone.

Other preferred embodiments of the present invention are provided by carrying out in addition a treatment with hydrogen peroxide and with UV radiation prior to, during or
25 after the treatment with ozone. The treatment with hydrogen peroxide and with UV radiation is preferably carried out at the same time as the treatment with ozone.

The invention is explained in the following with the aid of a drawing (Fig. 1) representing a preferred embodiment.

The water is brought to the required temperature by means of the heat exchanger 1. The water is passed through the adsorption column 2 only if the TOC content is unusually high. Appropriate valves are provided for this. Where the TOC content is low, the water can likewise be passed through the adsorber in order to desorb the TOC again. The appropriately heated water is mixed with hydrochloric acid 4 in the agitated tank 3 in order to adjust the pH value. The treatment with ozone then takes place in the columns 5 and 6. Because of the high potential corrosiveness of the ozone-oxygen mixture in water, both columns are preferably made of titanium. The water is delivered via the pump 7. The columns can be heated and are connected with one another by an overflow. The heat exchangers 8 and 9 serve to recondense water which may have been carried over (stripped) by the flow of gas. The ozone is produced from oxygen in the ozone generator 10 and added to the waste water via a nozzle 11 and 12 at the inlet to each of the columns. Unused ozone is thermally annihilated in the residual-ozone annihilator 13, preferably at a temperature of 250°C to 300°C.

For the analysis of the experiments, three sampling points 14 (blank test), 15 (downstream of the first column) and 16 (downstream of the second column) were installed for the withdrawal of the treated waste water. The ozone concentration is determined at the measuring points 17, 18, 19 and 20, downstream of the ozone generator and downstream of the heat exchangers 8 and 9.

The invention is illustrated in the following by Examples.

In the pilot plant reproduced in Fig.1, the waste water from the production of bisphenol A homopolycarbonate by the phase interface process was treated with different quantities of ozone at different temperatures and different pH values. The precise experimental conditions and results are shown in Table 1 (Examples according to the invention) and Table 2 (Comparison Examples).

The water supplied to the process contained 4 to 12 wt.% common salt and 0.3 to 1.5 wt.% carbonate. The TOC values of the water are given in the Tables. The TOC arose substantially from phenol and bisphenol A.

- 5 The experiments prove that, in order to decrease the TOC to values of approximately 1 ppm, it is particularly advantageous to establish an initial pH value of less than 7 and a temperature of between 60°C and 90°C. In the course of the reaction, a pH value of above 7 is established in column 1 and a pH value of about 8 is obtained after column 2. This change in the pH value is due to the fact that the CO₂ contained in the waste
- 10 water can escape only to some extent during the neutralisation and escapes only in the first column, so that an associated rise in the pH value takes place. This is accompanied by a change in the reaction mechanism from oxidation through attack by ozone to oxidation through attack by hydroxyl groups. The attack by ozone leads to the decomposition of the phenols and the formation of decomposition products, which are
- 15 then decomposed by the hydroxyl groups.

Table 1 (Examples according to the invention)

Waste water	Feed of ozone-containing gas to column 1	Feed of ozone-containing gas to column 2	Ozone-concentration in ozone-containing gas	TOC at 14	TOC at 15	TOC at 16	pH at 14	pH at 15	pH at 16	Temperature in column 1/ temperature in column 2
l/h	l/h	l/h	g/m ³	mg/l	mg/l	mg/l				°C
100	350	100	102	7.5	2.5	1.7	7.2	8.2	8.5	80/76
100	350	100	100	5	2.3	1.3	7.1	8.2	8.5	80/75
100	280	100	110	4.9	1.8	0.8	6.9	8.1	8.5	81/77
100	350	90	103	6.6	3.2	1.2	7	7.8	8.4	82/77
100	350	90	102	8.5	1.8	1.1	7	8.3	8.5	81/77
100	350	100	99.4	6.6	1.7	0.7	6.7	7.8	8.4	84/79
100	350	100	96	9.7	2	0.7	6.8	7.7	8.3	83/79
100	500	100	103	13.5	4.3	1.4	6.4	7.7	8.2	93/80
100	310	70	111	7.1	1.2	0.6	6.7	7.8	8.3	82/79
100	350	90	103	8.3	1.7	0.6	6.6	7.9	8.3	82/80
100	350	100	101	7	3.4	2.4	6.1	6.8	5.7	73/71
100	350	100	100	5.5	0.9	<0.5	6.3	6.9	6	74/71
100	350	100	101	8.2	1.9	0.6	6.4	6.9	6.1	74/70
100	350	100	102	5.4	1.2	0.9	4.6	6	6.3	95/76
100	350	100	99.4	4	1.4	0.6	6	6.4	6.4	55/59

Table 1 (Continuation)

Waste water	Feed of ozone-containing gas to column 1	Feed of ozone-containing gas to column 2	Ozone-concentration in ozone-containing gas	TOC at 14	TOC at 15	TOC at 16	pH at 14	pH at 15	pH at 16	Temperature in column 1/ temperature in column 2
l/h	l/h	l/h	g/m ³	mg/l	mg/l	mg/l				°C
100	350	100	101	5.7	1.1	<0.5	6	7.2	8.1	80/82
100	350	100	101	2.1	1	<0.5	6.1	6.9	7.9	70/76
100	350	100	105	11.3	5.2	1.2	6.3	6.9	7.8	71/74
100	350	100	99.6	5.4	2.2	1.4	6.3	7	7.9	73/76
100	350	100	100	9.8	1.6	0.9	6.3	6.7	6.8	73/77
100	350	100	102	3.1	2.5	0.5	5.6	6	6.4	73/65
100	350	100	99.4	11.3	1.8	0.7	6.1	6.5	7	91/91
100	350	100	127	10.7	1.5	<0.5	6.3	7.3	7.4	90/91
100	350	100	111	11.1	1.9	0.7	6.1	7	8.4	91/93
100	350	100	100	6.6	1.4	0.9	6.4	7.8	8.5	91/93
100	350	100	108	9.1	1.2	<0.5	6.4	7.8	8.5	90/91
100	350	100	102	10.6	1.5	<0.5	6.4	7.4	8.5	90/92

Table 2

(Comparison Examples; pH value of the water prior to ozone treatment: 12, temperature of the water prior to ozone treatment: 60°C)

Waste water	Feed of ozone-containing gas to column 1	Feed of ozone-containing gas to column 2	Ozone-concentration in ozone-containing gas	TOC at 14	TOC at 15	TOC at 16
l/h	l/h	l/h	g/m ³	mg/l	mg/l	mg/l
400	400	100	100	12.4	9.3	8.7
500	600	100	100	11.5	10	8.7
500	350	100	140	21.5	17.8	11.1
500	350	100	100	9.5	8.7	6.4
700	500	100	100	25.6	22.7	23.5
700	700	100	100	27.9	19.9	18.1

Claims

1. Process for the treatment, with ozone, of water which has a TOC of more than 2 ppm and contains at least 0.1 wt.% of dissolved carbonic acid or carbonates, characterised in that the treatment is carried out at a temperature of 10°C to 130°C and at an absolute pressure of 0.5 to 3 bar and that the pH value of the water which is supplied to the process is 2 to 11 and that the treatment takes place over a period of from 1 minute up to 10 hours.
2. Process according to claim 1, wherein the water contains 2 to 20 wt.% common salt.
3. Process for the production of chlorine by electrolysis of common salt, characterised in that the common salt is supplied to the electrolysis process in the form of an aqueous solution, which is obtained by the treatment, with ozone, of water which has a TOC of more than 2 ppm and a common salt content of 2 to 20 wt.% and contains at least 0.1 wt.% of dissolved carbonic acid or carbonates, wherein the treatment is carried out at a temperature of 10°C to 130°C and at an absolute pressure of 0.5 to 3 bar and wherein the pH value of the water which is supplied to the process for the treatment with ozone is 2 to 11 and wherein the treatment with ozone takes place over a period of from 1 minute up to 10 hours.
4. Process according to claim 3, wherein the electrolysis is carried out by the membrane process.
5. Process according to one of claims 1 to 4, wherein the water which is subjected to the treatment with ozone according to the invention is the waste water from the production of polycarbonate by the phase interface process.

6. Process according to claim 5, wherein the water which is subjected to the treatment with ozone according to the invention is the waste water from the production of bisphenol A polycarbonate by the phase interface process.
- 5 7. Process according to one of claims 1 to 6, wherein the pH of the water supplied to the process for the treatment with ozone has a value which is less than 7 and is such that, after the treatment of the water with ozone, the pH value is more than 7.5.

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Fig. 1

