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(54) **Title:** NOVEL METHOD OF DOSING LIQUIDS WITH PRESSURIZED OZONE

(57) **Abstract:** Methods of treating and sanitizing foods and other targeted objects, including the use of ozone and a method for providing a liquid dosed with pressurized ozone. The ozonated liquid of the invention is particularly useful for treating food products, food storage, and food transportation devices as well as treating water, or other target objects. Included is a method or pressurizing an ozone-containing stream without destroying the ozone or contaminating the stream with oil or water. The pressurized ozone-containing stream is then used to dose a liquid, which in turn is used to sanitize or treat a target object.

NOVEL METHOD OF DOSING LIQUIDS WITH PRESSURIZED OZONE

Background

Treating and sanitation of food, equipment, pharmaceutical products, and even water to reduce undesirable biological microorganisms is important to the protection of public health. For example, food can be damaged by microbes, spores, insects, and other similar sources. Each year, economic losses of food and labor due to damage from such sources is more than $100 billion. Currently, food items are preserved using a variety of methods, including refrigeration, fumigation with toxic chemicals, irradiation, biological control, heat exposure, and controlled atmosphere storage (a fruit industry technique that involves modifying the concentration of gases naturally present in the air).

The primary problem regarding food spoilage in public health is microbial growth. If pathogenic microorganisms are present, then growth can potentially lead to food-borne outbreaks and significant economic losses. Food safety concerns have been brought to the consumers' attention since the early part of the 20th century and those concerns have become even stronger today. Outbreaks from *Salmonella* and *E. coli* have increased the focus on food safety, including from a regulatory perspective. A study completed by the Centers for Disease Control and Prevention (CDC) estimated that food-borne diseases cause approximately 76 million illnesses, 325,000 hospitalizations, and 5,000 deaths annually in the US. Those numbers reveal the dramatic need for effective means of handling food products in order to ensure food safety.

Effective sanitation of food or other items depends on the combination of what is to be sanitized and the sanitation process type. Not all of the currently available technologies can deliver an effective reduction of microorganisms and at the same time prevent product or environmental degradation. It is well known in the art to cool products, such as foods, during processing with some type of refrigerant to slow down the growth of unwanted microbes and enzymatic reactions in foods. For instance, the shelf life and quality of food products are improved by processing, transporting, and storing under refrigerated conditions.

Cooling agents, such as water ice, dry ice, carbon dioxide, or nitrogen, are liquid or solid agents that can be used as an expendable refrigerant. In food processing applications, liquids, such as nitrogen, are used to cool and inert the atmosphere during food processing or storage.

While refrigeration can retard microbial growth, such treatment does not necessarily kill bacteria. Accordingly, microorganisms can still survive through refrigeration, and worse, some microorganisms can still grow and produce harmful substances during refrigerated storage. Furthermore, it is possible that the refrigerant used to cool a target item or food product can itself be contaminated with pathogenic microorganisms, thus contaminating the target item or food product.

Biocidal agents are used to sanitize equipment, provide antiseptic environments, treat water, and sanitize foods. The reaction of biocidal agents with microbial cell structures is often irreversible; therefore the cells either become attenuated or die.

One biocidal agent commonly used in the industry is ozone. However, ozone is very unstable, and therefore, must be produced at the location of consumption. Production of ozone requires specialized equipment and involves safety issues due to handling of the equipment and feedstock, such as pure oxygen. After the ozone is produced, it must be delivered in some form to the target item as a sanitizer. Ozone is often dissolved or absorbed in water as a mechanism to deliver the unstable ozone to a target item. However, ozone has poor solubility in water. Mixtures of ozone and water typically contain less than about 20 ppm by weight (ppmwt) ozone. As a result, large quantities of water relative to the ozone are required if water is used as a delivery agent. Furthermore, because of the large quantities of water required, the ozone and water cannot be pre-mixed and transported to site. Thus, ozone and water must be mixed on site.

Another problem with ozone is the difficulty in compressing an ozone-containing stream. There are no commercial processes known to one of ordinary skill in the art that are capable of delivering ozone at high pressures.

Ozone generating equipment known in the art typically produces an ozone-containing gas stream at fairly low pressures. These ozone generators are typically limited to producing a stream with a pressure of less than about 25 psig. Conventional mechanical compression cannot be used to compress ozone because the unstable ozone molecule is destroyed in conventional compressors. Oil-lubricated or water ring compression can be used to compress a stream containing ozone up to 150 psig; however, these compressors inherently contaminate the ozone stream with oil or water respectively. Therefore, the prior art fails to provide a method to compress the ozone to pressures above about 25 psig without contaminating the ozone stream with some level of oil or water. Furthermore, the prior art fails to provide any method to successfully compress an ozone stream to pressures of greater than about 150 psig without destroying the ozone.

It is desirable to pressurize ozone to be used to sanitize equipment or devices and process foods. It is particularly desirable to be able to provide ozone in a pressurized stream at pressures above 150 psig without contaminating the ozone with oil or water. Further yet, it is desirable to pressurize ozone and feed it into a liquid, so that it is absorbed under pressure into the liquid and the liquid can them be used to treat of sanitize devices or food products.

Summary

The current invention fulfills the need to provide a process to pressurize an ozone-containing stream without destroying the ozone or contaminating the ozone-containing stream with oil or water. It is desirable to be able to provide a pressurized ozone-containing stream that is substantially free of oil and moisture. It is also desirable to the ozone-containing stream at pressures above 150 psig. It further desirable to pressurize ozone and feed it into a liquid, so that it is absorbed under pressure into the liquid.

The current invention pressurizes ozone by feeding an ozone-containing source to an ozone pressurization system to establish a first pressure followed by pressurizing the ozone pressurization system. The ozone pressurization system is pressurized by feeding a pressurization gas to

the ozone pressurization system to raise the pressure to the second pressure and form a pressurized ozone-containing gas. The pressurized ozone-containing gas is then withdrawn from the ozone pressurization system. This method pressurizes the ozone-containing stream without contaminating the stream with oil or water.

In one embodiment, the ozone pressurization system comprises a pressurization vessel, the pressurization gas is fed into the lower portion of the pressurization vessel, and the pressurized ozone-containing gas is withdrawn from the upper portion of the pressurization vessel.

In another embodiment, the pressurization system comprises a first pressurization vessel and a last pressurization vessel. In this embodiment, the pressurization vessels are fluidly connected in series and the pressurizing gas is fed exclusively to the first pressurization vessel.

The current inventive method also provides a method to produce an ozonated liquid by feeding an ozone-containing gas from the ozone source to an ozone pressurization system to establish a first pressure, pressurizing the ozone pressurization system by feeding a pressurization gas into the system; thus, raising the pressure of the ozone-containing gas to a second pressure and forming a pressurized ozone-containing gas. This method also places a liquefied dry gas in an expansion vessel, and sparges the pressurized ozone-containing gas through the liquefied dry gas to form an ozonated liquefied dry gas, which is withdrawn from the expansion vessel.

Brief Description of the Drawings

For a further understanding of the nature and objects of the present invention, reference should be made to the following detailed description, taken in conjunction with the accompanying drawings, in which like elements are given the same or analogous reference numbers and wherein:

- Figure 1 is a schematic of an embodiment of the current invention for pressurizing ozone;
- Figure 2 is a schematic of another embodiment of the current invention for pressurizing ozone;

- Figure 3 is a schematic of yet another embodiment of the current invention for pressurizing ozone;
- Figure 4 is a schematic of still another embodiment of the current invention for pressurizing ozone;
- Figure 5 is a schematic of an embodiment for pressurizing ozone and producing an ozonated liquefied gas according to the current invention; and
- Figure 6 is a graph showing the concentrations of ozone in a liquefied gas attained by the current method of the current invention.

Description of Preferred Embodiments

The current invention provides a method of pressurizing an ozone-containing stream without destroying the ozone or contaminating the ozone-containing stream with oil or water. Furthermore, the method forms an ozonated liquefied dry gas by bubbling the pressurized ozone-containing stream through a reservoir of liquefied dry gas.

The current invention pressurizes ozone by feeding an ozone-containing source to an ozone pressurization system to establish a first pressure followed by pressurizing the ozone pressurization system. The ozone pressurization system is pressurized by feeding a pressurization gas to the ozone pressurization system to raise the pressure to the second pressure and form a pressurized ozone-containing gas. The pressurized ozone-containing gas is then withdrawn from the ozone pressurization system.

In one preferred method, the pressurization gas is a dry gas. One preferred dry gas is CO_2. The method pressurizes the ozone-containing stream without contaminating the stream with oil or water. The method preferable pressurizes the ozone-containing stream to a pressure that is greater than about 150 psig. Furthermore, the method can pressurize the ozone-containing stream from a pressure of less than about 50 psig to a pressure that is preferably greater than about 150 psig and even more preferably greater than about 200 psig.

In one embodiment of the method, the ozone pressurization system comprises a pressurization vessel, the pressurization gas is fed into the lower portion of the pressurization vessel, and the pressurized ozone-containing gas is withdrawn from the upper portion of the pressurization vessel.

In another embodiment, the pressurization system comprises a first pressurization vessel and a last pressurization vessel. In this embodiment, the pressurization vessels are fluidly connected in series and the pressurizing gas is fed exclusively to the first pressurization vessel. In a preferred embodiment, the pressurized gas mixture is withdrawn from the first pressurization vessel and fed to the last pressurization vessel, while the pressurized ozone-containing gas is withdrawn from the last pressurization vessel. In a further preferred embodiment, the pressurization gas is fed into a lower portion of the first pressurization vessel while the pressurized gas mixture is withdrawn from an upper portion of that first pressurization vessel. The pressurized gas mixture is then fed into a lower portion of the last pressurization vessel and the pressurized ozone-containing gas is finally withdrawn from an upper portion of the last pressurization vessel.

In yet a further embodiment, the pressurization system comprises a number of pressurization vessels in series and includes the steps of withdrawing a pressurized gas mixture from the first pressurization vessel, feeding the pressurized gas mixture to a successive pressurization vessel, and transferring the pressurized gas mixture from the successive pressurization vessel to the last pressurization vessel. This embodiment, like those above withdraws the pressurized ozone-containing gas from the last pressurization vessel. One further embodiment of this method feeds the pressurization gas into a lower portion of the first pressurization vessel, withdraws the pressurized gas is from an upper portion of the first pressurization vessel, feeds the pressurized gas mixture into a lower portion of the successive pressurization vessel, withdraws that pressurized gas from an upper portion of the successive pressurization vessel, and ultimately transfers the pressurized gas mixture into a lower portion of the last pressurization vessel.

The current inventive method also provides an ozonated liquid by feeding an ozone-containing gas from the ozone source to an ozone pressurization system to establish a first pressure, pressurizing the ozone pressurization system by feeding a pressurization gas into the system thus raising the pressure of the ozone-containing gas to a second pressure and forming a pressurized ozone-containing gas. The method also places a liquefied dry gas in an expansion vessel, and sparges the pressurized ozone-containing gas through the liquefied dry gas to form an ozonated liquefied dry gas, which is withdrawn from the expansion vessel. This method may further include steps of withdrawing a vent gas from the expansion tank and recycling the vent gas to a liquefied dry gas storage vessel. In some embodiments, the sparging occurs substantially continuously.

Referring to Figure 1, one embodiment of the current method provides an ozone-containing gas **102** from an ozone source **104**. Ozone can be generated in commercially available ozone generator known to one of ordinary skill in the art. The ozone generation unit preferably uses a pure oxygen feed, to form the ozone-containing gas **102**, which preferably contains about 6 to about 13 wt% ozone in oxygen, and more preferably about 9 to about 11 wt% ozone. The ozone pressurization system **106** is purged with the ozone-containing gas **102** to establish a concentration of ozone in the entire ozone pressurization system **106**. Then, the ozone pressurization system **106** is pressurized with the ozone-containing gas **102** to set an initial pressure, for instance about 5 to about 25 psig, in the entire ozone pressurization system **106**. The higher the initial pressure in the compression system, the higher final pressure that can be achieved. The entire ozone pressurization system **106** is then isolated from the ozone generator. The ozone-containing gas **102** is pressurized in an ozone pressurization system **106** to form a pressurized ozone-containing gas **108**. To form the pressurized ozone-containing gas **108**, the ozone-containing gas **102** is fed to the ozone pressurization system **106** to establish a concentration of ozone-containing gas throughout the ozone pressurization system **106** at first pressure in the ozone pressurization system **106**. The first pressure is preferably less than about 100 psig, more preferably less than about 50 psig and even more preferably less than about 30 psig. Next, a pressurization gas **110** is fed to the ozone pressurization system **106** to raise the pressure in the ozone pressurization system **106** to a second pressure. As the pressurization gas **110** enters the ozone pressurization system **106**, the ozone-containing gas in the ozone pressurization system **106** is compressed. It is believed that if the density of the pressurization gas **110** and the ozone-containing gas **102** is substantially different, the gases stratify in the tanks and mixing is minimal. The final result is a compressed ozone-containing feed mixture typically containing close to, but somewhat lower concentration of ozone in oxygen that the ozone-containing gas **102** in the upper portion of any vessels in the ozone pressurization system **106**. The pressurization gas **110** feed to the ozone pressurization system **106** is stopped when the desired second pressure is reached. The pressurized ozone-containing gas **108** is then withdrawn from the ozone pressurization system **106**. The ozone pressurization system **106** preferably comprises at least one pressurization vessel **112**. Furthermore, the pressurization gas **110** is preferably fed to a lower portion of the pressurization vessel **112** and more preferably fed to the bottom portion of the pressurization vessel **112**. The pressurized ozone-containing gas **108** is preferably removed from an upper portion of the pressurization vessel **112** and more preferably removed from the top of the pressurization vessel **112**. In one preferred embodiment, the pressurization gas **110** is fed at a slow flow rate to the pressurization system **106** in order to minimize the mixing for the ozone-containing gas **102** and the pressurization gas **110**. It is preferable to maintain laminar flow rates in the pressurization vessel **112**.

Still referring to Figure 1, the pressurization gas **110** is fed to the pressurization vessel to raise the pressure in the ozone pressurization system **106** to a second pressure of greater than about 100 psig, more preferably greater than about 150 psig, and even more preferably greater than about 200 psig. Then, the pressurized ozone-containing gas **108** can be withdrawn from the ozone pressurization system **106**. Using this method, it is feasible to pressurize an ozone-containing stream to pressures of greater than about 500 psig and even greater than about 1,000 psig without destroying the ozone.

Furthermore, a substantial portion of the ozone (O_3) remains relatively undiluted. Still further, the resulting pressurized ozone-containing gas **108** is substantially free of oil, water, or other undesirable contaminants. In one preferred embodiment, the pressurized ozone-containing gas **108** contains less than about 0.05 wt% water, preferably contains less than about 200 ppm wt water, and more preferably less than about 20 ppm wt water.

One preferred pressurization gas **110** is a dry gas. The dry gas can be any suitable non-aqueous gas, but is preferably a liquefied gas, particularly a liquefied gas with a high gas density compared to the ozone-containing gas **102**. The dry gas preferably contains less than 0.05 wt% water, and more preferably containing less than 20 ppm wt water. In one preferred embodiment, the pressurization gas **110** is preferably a dry gas that is stored as a liquid, such as CO_2. The liquefied dry gas is removed from the liquid storage vessel and expanded to form the pressurization gas **110**. One preferred embodiment uses a pressurization gas **110** that has a gas density that is higher that the gas density of the ozone-containing gas **102**. This pressurization gas **110** is preferably cold after expansion to provide a pressurization gas at the highest gas density possible for that gas. In one embodiment, the pressurization gas is preferably less than about 20°C after expansion, and more preferably less than about 10°C after expansion. By using a pressurization gas **110** that is higher in gas density than the ozone-containing gas **102**, and by feeding the pressurization gas **110** slowly, dilution of the ozone-containing gas **102** with the pressurization gas **110** is minimized. Without being limited by this theory, it is believed that the higher gas density pressurization gas **110** layers out in the pressurization vessel **112** below the lower density ozone-containing gas **102** already in the vessel. In one embodiment, the concentration of ozone in the pressurized ozone-containing gas **108** is at least about 70% of the concentration of ozone in the ozone-containing gas **102**, more preferably at least 80% of the concentration of ozone in the ozone-containing gas **102**, and even more preferably at least about 90% of the concentration of ozone in the ozone-containing gas **102**.

In one embodiment, a continuous flow of pressurized ozone gas **110** is supplied by using a plurality of ozone pressurization systems **106** operated in a “round robin” to maximize the use of the ozone generator and minimize the waste of pressurization gas **110** by cross-tying (not shown) the sets of ozone pressurization systems **106**.

Referring to Figure 2, one embodiment of the current method utilizes an ozone pressurization system **106** that comprises a plurality of pressurization vessels, which comprise at least a first pressurization vessel **202** and a last pressurization vessel **204**. An upper portion of the first pressurization vessel **202** and a lower portion of the last pressurization vessel **204** are connected by a fluid connection means **206**. The fluid connection means can be any type or combination of pipe, conduit, vessel, valve, orifice, chamber, or other flow passage that allows a pressurized gas mixture to flow from one vessel to another. One preferred fluid connection means **206** has at least part of the means that is smaller in diameter than the first pressurization vessel **202** or the last pressurization vessel **204**. In this preferred method, a pressurized gas mixture is withdrawn (or pushed) from the upper portion of the first pressurization vessel **202** and fed into a lower portion of the last pressurization vessel **204** as the pressurization gas **110** is fed into the lower portion of the first pressurization vessel **202**. Eventually, the first pressurization vessel **202** will be completely filled with a gas mixture that primarily comprises the pressurization gas **110**. Having a plurality of pressurization vessels helps prevent the pressurization gas **110** from mixing with the ozone-containing gas **102**. The compressed ozone-containing gas **110** is withdrawn from the last pressurization vessel **204**. The last pressurization vessel **204** is preferably isolated from any upstream pressurization vessels to prevent unwanted dilution of the pressurized ozone mixture. The pressure from the last pressurization vessel **204** is allowed to drop as the ozone-containing gas **110** is fed to the process.

Referring to Figure 3, one embodiment of the current method uses a plurality of pressurization vessels comprising more than two pressurization vessels fluidly connected in series. The ozone-containing gas **102** is first fed to fill the plurality of pressurization vessels with the ozone-containing gas **102** at a first pressure. Next, the pressurization gas **110** is fed into a first pressurization vessel **202** in the series of pressurization vessels and the pressurized ozone-containing gas **108** flows out of a last pressurization vessel **204** in the series of pressurization vessels. The embodiment of Figure 3 uses an ozone pressurization system that comprises a first pressurization vessel **202**, a successive pressurization vessel **303,** and a last pressurization vessel **204**. In this embodiment, a pressurized gas mixture is withdrawn from the first pressurization vessel **202** and fed to the successive pressurization vessel. The pressurized gas mixture then flows from the successive pressurization vessel to the last pressurization vessel **204**. The pressurized ozone-containing gas **108** is then withdrawn from the last pressurization vessel **204** after the ozone pressurization system **106** reaches a second pressure.

Still referring to Figure 3, in one embodiment, the pressurization gas is fed into a lower portion of the first pressurization vessel **202**. A pressurized gas mixture is withdrawn from an upper portion of the first pressurization vessel **202** and fed into a lower portion of the successive pressurization vessel **302** via a first conduit **304**. The pressurized gas is then withdrawn from an upper portion of the successive pressurization vessel **302** and transferred into a lower portion of the last pressurization vessel **204** via a second conduit **306**. Finally, the pressurized ozone-containing gas **108** is withdrawn from the upper portion of the last pressurization vessel **204** after the ozone pressurization system **106** reaches a second pressure.

Referring to Figure 4, one embodiment of the current method uses a plurality of pressurization vessels fluidly connected in series. In this embodiment, the ozone pressurization system **106** comprises a first pressurization vessel **202**, at least two successive pressurization vessels **402, 404**, and a last pressurization vessel **204**, all fluidly connected in series. The ozone-containing gas **102** is first fed to fill all of the pressurization vessels **202, 204, 402, 404** with the ozone-containing gas **102** at a first pressure. The pressurization gas **110** is fed into a first pressurization vessel **202** and the pressurized ozone-containing gas **108** flows out of a last pressurization vessel **204**. In this embodiment, a pressurized gas mixture is withdrawn from the first pressurization vessel **202** and fed to the successive pressurization vessels

402 and **404** in series via a plurality of gas transfer conduits **406, 408, 410**. The pressurized gas mixture then flows from the successive pressurization vessels **402** and **404** to the last pressurization vessel **204**. The pressurized ozone-containing gas **108** is then withdrawn from the last pressurization vessel **204** after the ozone pressurization system **106** reaches a second pressure. As described above, in a preferred embodiment, the incoming gases flow into the lower portions of the respective vessels and the outgoing gases exit the upper portions of the respective vessels.

In one embodiment of the current invention, a continuous supply of pressurized ozone feed is supplied. In the embodiment, the pressurization vessels upstream of the last pressurization vessel **204** are replenished with ozone-containing gas **102**. To accomplish this, the tanks upstream of the last pressurization vessel **204** are vented of their pressure, purged, re-filled with the ozone-containing gas **102**, and re-pressurized as described above. This new batch of pressurized gas may then be released into the last pressurization vessel **204**. This re-filling gives a slightly more dilute ozone mixture. A more efficient arrangement consists of several sets of tanks, operated in a “round robin” to maximize the use of the ozone generator, capture all pressurized ozone that does not reach the last pressurization vessel **204**, and minimize the waste of pressurization gas **110** by allowing the sets of tanks to be cross-tied.

Now referring to Figure 5, the current inventive method also provides an ozonated liquefied dry gas **502** by transferring a liquefied dry gas **504** to an expansion vessel **506** and sparging the pressurized ozone-containing gas **110** through the liquefied dry gas to form the ozonated liquefied dry gas **502**. In one embodiment of this method, the liquefied dry gas **504** is stored in a liquefied dry gas storage vessel **508** at a pressure suitable to maintain the dry gas in liquid form. The liquefied dry gas **504** is transferred to the expansion vessel **506** where the pressure is somewhat less than the pressure in the liquefied dry gas storage vessel **508**. The ozone pressurization system **106** pressurizes an ozone-containing gas **102** as described in the previous embodiments to form a pressurized ozone-containing gas **110** at a second pressure, which is above the pressure in the expansion vessel **506**. The second pressure is preferably at least about 50 psig above the pressure in the expansion vessel **506**, and more preferably at least about 100 psig above the pressure in the expansion vessel **506**. The pressurized ozone-containing gas **110** is then sparged through the liquefied dry gas in the expansion vessel **506** to form the ozonated liquefied dry gas **502**. The ozonated liquefied dry gas is withdrawn from the expansion vessel **506** after sufficient sparging to assure the liquid contains a desired amount of ozone. In one embodiment, the expansion vessel **506** is substantially filled with the liquefied dry gas **504** and then the pressurized ozone-containing gas **110** is sparged through the liquid on a batch basis. In other embodiments, there is a continuous flow of liquefied dry gas **504** into the expansion vessel **506** and the pressurized ozone-containing gas **110** is sparged continuously through the liquid. This method may further include steps of withdrawing a vent gas **510** from the expansion tank. In some embodiments, the vent gas **510** is fed to a recovery system **512** for recovery and recycling of the vent gas back to the liquefied dry gas storage vessel

Still referring to Figure 5, one embodiment of the current invention uses CO_2 as the liquefied dry gas. In this embodiment, the pressure in the liquefied dry gas storage vessel **508** is about 200 to about 400 psig. The liquefied dry gas **504** is transferred to the expansion vessel **506** where the pressure is preferably about 100 to about 200 psig, more preferably about 150 to 200 psig, and even more preferably about 200 to about 300 psig. The ozone pressurization system **106** provides the pressurized ozone-containing gas **110** at a second pressure, which is above the pressure in the expansion vessel **506**, preferably at least about 50 psig above the pressure in the expansion vessel **506**, and more preferably at least about 100 psig above the pressure in the expansion vessel **506**. The ozonated liquefied dry gas is withdrawn from the expansion vessel **506** after sufficient sparging to assure the liquid CO_2 contains a desired amount of ozone. By this method, an ozonated liquid CO_2 can be supplied containing at least about 200 ppm wt ozone, and more preferably greater than about 250 ppm wt ozone. As is shown in Figure 5, the current method provides an ozonated liquid with a much higher concentration of ozone that the prior art methods of saturating water with ozone.

Another embodiment of the dry gas pressurization method described above is used to dose other liquids, including aqueous and dry (non-aqueous) liquids. In one embodiment of the invention, the dry gas compression method is used to ozonate a liquid, where the liquid is at pressures greater than about 150 psig. In this embodiment, a liquid is placed into a pressure vessel where the pressure is greater than about 150 (or is raised to above this pressure), preferably great than about 200 psig, and more preferably greater than about 300 psig. The ozone pressurization system provides the pressurized ozone-containing gas at a second pressure, which is above the pressure in the pressure vessel, preferably at least about 50 psig above the pressure in the expansion vessel, and more preferably at least about 100 psig above the pressure in the expansion vessel. The ozonated liquid is withdrawn from the pressure vessel after sufficient sparging to assure the liquid contains a desired amount of ozone.

Although the present invention has been described in considerable detail with reference to certain preferred versions and examples thereof, other versions are possible. For instance, any liquid which one skilled in the art wishes to saturate with ozone can be substituted for the liquefied dry gas of the current invention in the method. Furthermore, there is a large variety of configurations of vessels, pipes, and other equipment that can be used as pressurization vessels. Clearly, the current invention may be used in a variety of processes for processing food, or non-food items. Therefore, the spirit and scope of the appended claims should not be limited to the description of the preferred versions contained herein.

All the features disclosed in this specification (including any accompanying claims, abstract, and drawings) may be replaced by alternative features serving the same, equivalent or similar purpose, unless expressly stated otherwise. Thus, unless expressly stated otherwise, each feature disclosed is one example only of a generic series of equivalent or similar features.

CLAIMS:

1. A method of pressurizing ozone comprising the steps of:
   a) providing an ozone source and an ozone pressurization system;
   b) feeding an ozone-containing gas from said ozone source to said ozone pressurization system to establish a first pressure;
   c) pressurizing said ozone pressurization system to a second pressure by feeding a pressurization gas to said ozone pressurization system to form a pressurized ozone-containing gas; and
   d) withdrawing said pressurized ozone-containing gas from said ozone pressurization system.

2. The method of claim 1, wherein said pressurized ozone-containing stream is substantially absent oil and water.

3. The method of claim 1, wherein:
   a) said ozone pressurization system comprises a pressurization vessel;
   b) said pressurization gas is fed into the lower portion of said pressurization vessel; and
   c) said pressurized ozone-containing gas is withdrawn from the upper portion of said pressurization vessel.

4. The method of claim 1, wherein:
   a) said ozone pressurization system comprises a plurality of pressurization vessels comprising a first pressurization vessel and a last pressurization vessel;
   b) said plurality of pressurization vessels are fluidly connected in series; and

c) wherein said pressurizing gas is fed exclusively to said first pressurization vessel.

5. The method of claim 4, wherein said pressurizing step further comprises the steps of:

a) withdrawing a pressurized gas mixture from said first pressurization vessel;

b) feeding said pressurized gas mixture to said last pressurization vessel, wherein pressurized ozone-containing gas is withdrawn from said last pressurization vessel.

6. The method of claim 5, wherein:

a) said pressurization gas is fed into a lower portion of said first pressurization vessel;

b) said pressurized gas mixture is withdrawn from an upper portion of said first pressurization vessel;

c) said pressurized gas mixture is fed into a lower portion of said last pressurization vessel; and

d) said pressurized ozone-containing gas is withdrawn from an upper portion of said last pressurization vessel.

7. The method of claim 4, wherein said pressurizing step further comprises the steps of:

a) withdrawing a pressurized gas mixture from said first pressurization vessel;

b) feeding said pressurized gas mixture to a successive pressurization vessel; and

c) transferring said pressurized gas mixture from said successive pressurization vessel to said last pressurization vessel, wherein pressurized ozone-containing gas is withdrawn from said last pressurization vessel.

8. The method of claim 7, wherein:

a) said pressurization gas is fed into a lower portion of said first pressurization vessel to form a pressurized gas mixture;

b) said pressurized gas mixture is withdrawn from an upper portion of said first pressurization vessel;

c) said pressurized gas mixture is fed into a lower portion of said successive pressurization vessel;

d) said pressurized gas is withdrawn from an upper portion of said successive pressurization vessel; and

e) said pressurized gas mixture is transferred into a lower portion of said last pressurization vessel.

9. The method of claim 1, wherein said pressurization gas is a dry gas.

10. The method of claim 9, wherein said dry gas is CO_2.

11. The method of claim 1, wherein said second pressure is greater than about 150 psig.

12. The method of claim 11, wherein said first pressure is less than about 50 psig.

13. A method of supplying an ozonated liquid comprising the steps of:

a) providing an ozone source and an ozone pressurization system;

b) feeding an ozone-containing gas from said ozone source to said ozone pressurization system to establish a first pressure;

c) pressurizing said ozone pressurization system by feeding a pressurization gas into said ozone pressurization system, wherein said pressurizing gas raises the pressure of said ozone-containing gas to a second pressure to form a pressurized ozone-containing gas;

d) providing a liquefied dry gas;

e) transferring said liquefied dry gas to an expansion vessel;

f) sparging said pressurized ozone-containing gas through said liquefied dry gas in said expansion tank to form an ozonated liquefied dry gas; and

g) withdrawing said ozonated liquefied dry gas from said expansion vessel.

14. The method of claim 13, further comprising the steps of withdrawing a vent gas from said expansion tank and recycling said vent gas to a liquefied dry gas storage vessel.

15. The method of claim 13, wherein said sparging occurs substantially continuously.

16. The method of claim 13, wherein said pressurizing step further comprises the steps of:

a) providing a plurality of pressurization vessels fluidly connected in series in said pressurization system;

b) feeding said pressurizing gas exclusively to a first pressurization vessel in said pressurization system;

c) withdrawing a pressurized gas mixture from said first pressurization vessel;

d) feeding said pressurized gas mixture to said last pressurization vessel, wherein pressurized ozone-containing gas is withdrawn from said last pressurization vessel.

17. The method of claim 13, wherein said pressurized ozone-containing gas is substantially absent water and oil.

18. A device for supplying a pressurized ozone-containing gas comprising:

a) an ozone pressurization system comprising:

i) a first pressurization vessel;

ii) a last pressurization vessel;

iii) a pressurization inlet located on a lower portion of said first pressurization vessel;

iv) a means of fluidly connecting the upper portion of said first pressurization vessel to a lower portion of said last pressurization vessel; and

v) a pressurized ozone outlet located on an upper portion of said last pressurization vessel;

b) an ozone-containing feedstream in fluid communication with said first pressurization vessel;

c) a pressurization gas feedstream in fluid communication with said pressurization inlet; and

d) a pressurized ozone-containing stream in fluid communication with said pressurized ozone outlet.

19. The device of claim 18, further comprising a successive pressurization vessel, wherein:

a) an upper portion of said first pressurization vessel is fluidly connected to a lower portion of said successive pressurization vessel; and

b) an upper portion of said successive pressurization vessel is fluidly connected to a lower portion of said last pressurization vessel.

20. The device of claim 19, wherein said successive pressurization vessel comprises at least two further successive pressurization vessels fluidly connected in series.

21. The device of claim 18, wherein said pressurization gas feedstream comprises a dry gas.

22. The device of claim 21, wherein said dry gas is CO_2.

23. A method of supplying an ozonated liquid comprising the steps of:

a) providing an ozone source and an ozone pressurization system;

b) feeding an ozone-containing gas from said ozone source to said ozone pressurization system to establish a first pressure;

c) pressurizing said ozone pressurization system by feeding a pressurization gas into said ozone pressurization system, wherein said pressurizing gas raises the pressure of said ozone-containing gas to a second pressure to form a pressurized ozone-containing gas;

d) providing a liquid in a pressure vessel;

e) sparging said pressurized ozone-containing gas through said liquid in said pressure vessel to form an ozonated liquid; and

g) withdrawing said ozonated liquid from said pressure vessel.

24. The method of claim 23, wherein a pressure in said pressure vessel is greater than about 150 psig during said sparging step.

25. The method of claim 23, wherein a pressure in said pressure vessel is greater than about 200 psig during said sparging step.

26. The method of claim 23, wherein a pressure in said pressure vessel is greater than about 300 psig during said sparging step.

27. The method of claim 23, wherein said sparging occurs substantially continuously.

28. The method of claim 23, wherein said pressurizing step further comprises the steps of:

a) providing a plurality of pressurization vessels fluidly connected in series in said pressurization system;

b) feeding said pressurizing gas exclusively to a first pressurization vessel in said pressurization system;

c) withdrawing a pressurized gas mixture from said first pressurization vessel; and

d) feeding said pressurized gas mixture to said last pressurization vessel, wherein pressurized ozone-containing gas is withdrawn from said last pressurization vessel.

29. The method of claim 23, wherein said pressurized ozone-containing gas is substantially absent water and oil.

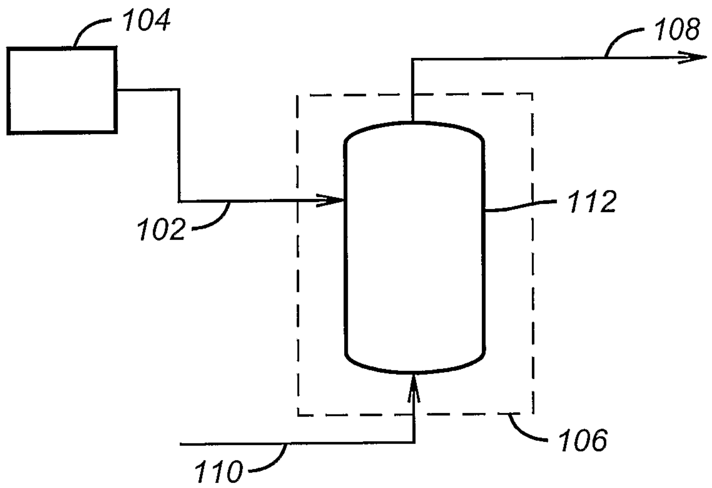

FIG. 1

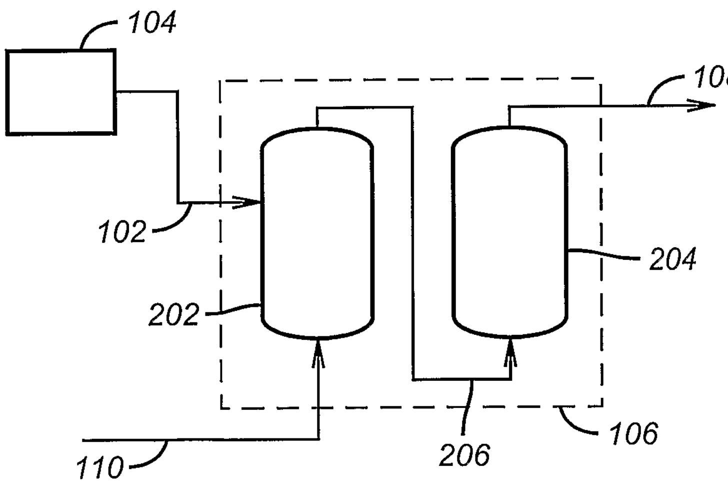

FIG. 2

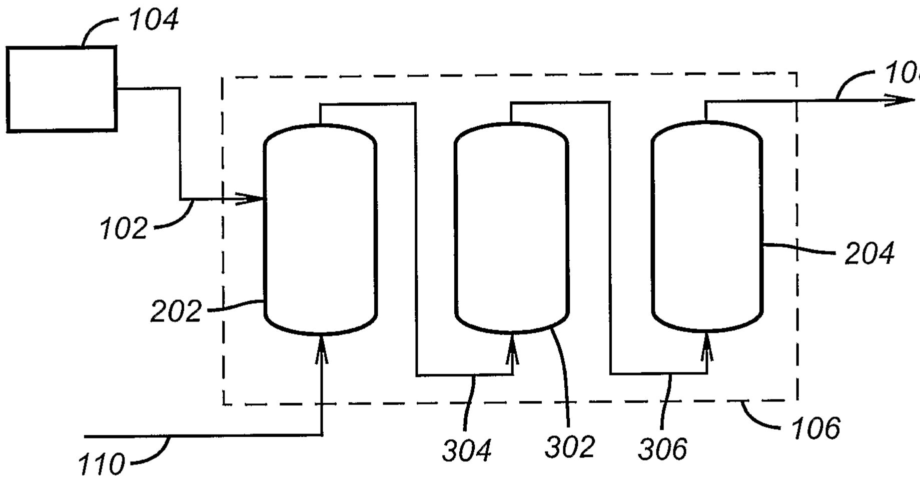
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FIG. 3

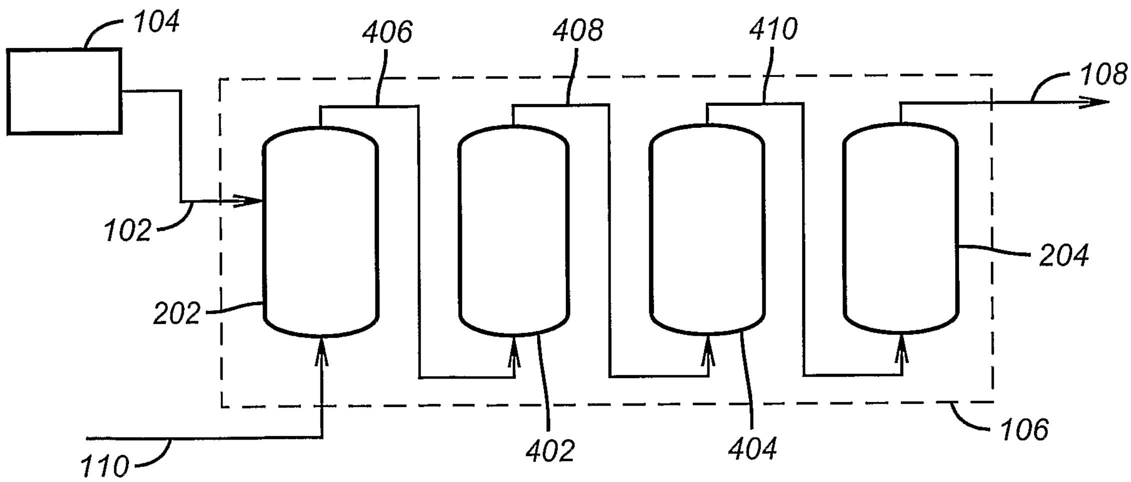
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FIG. 4

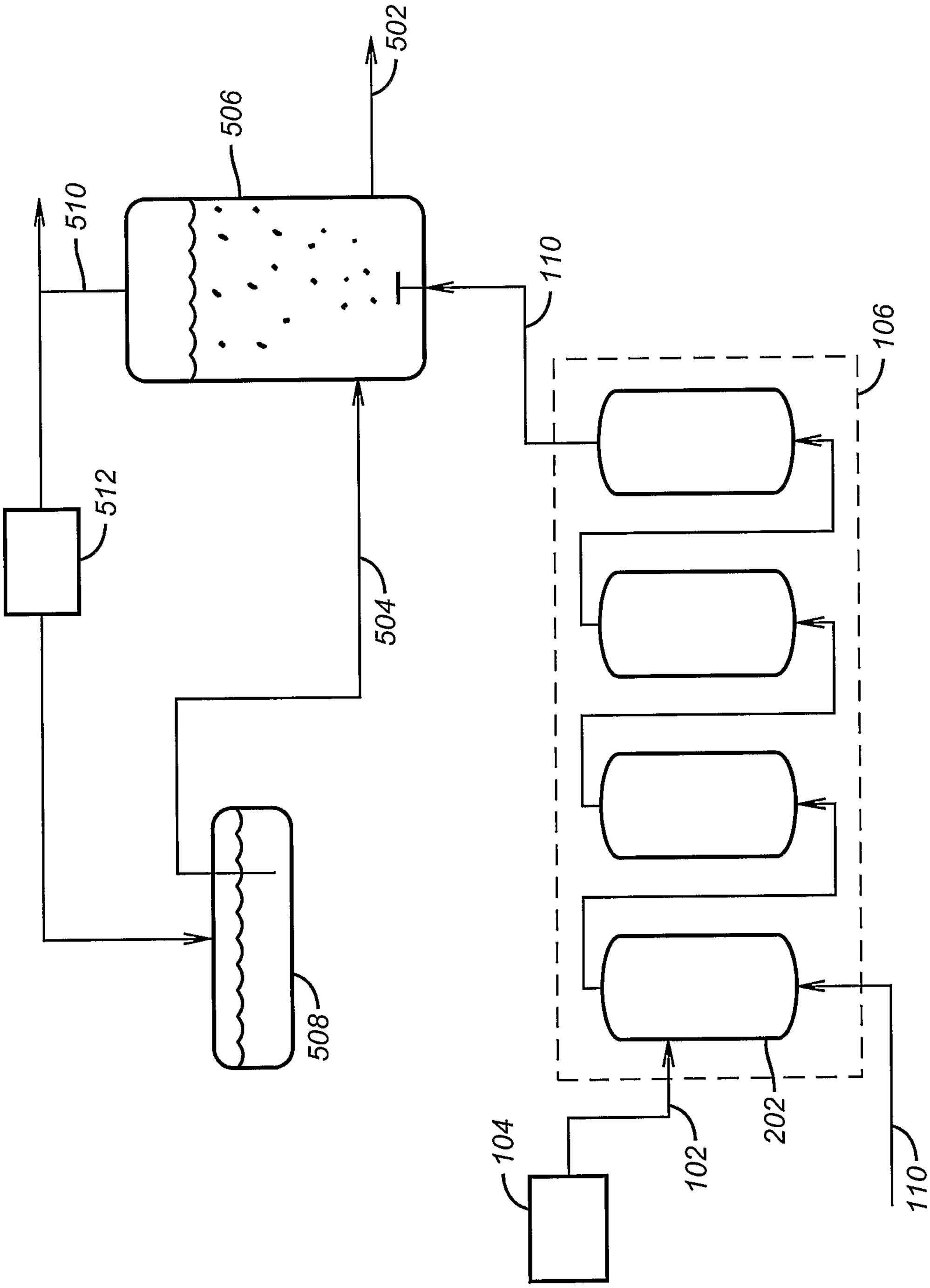

FIG. 5

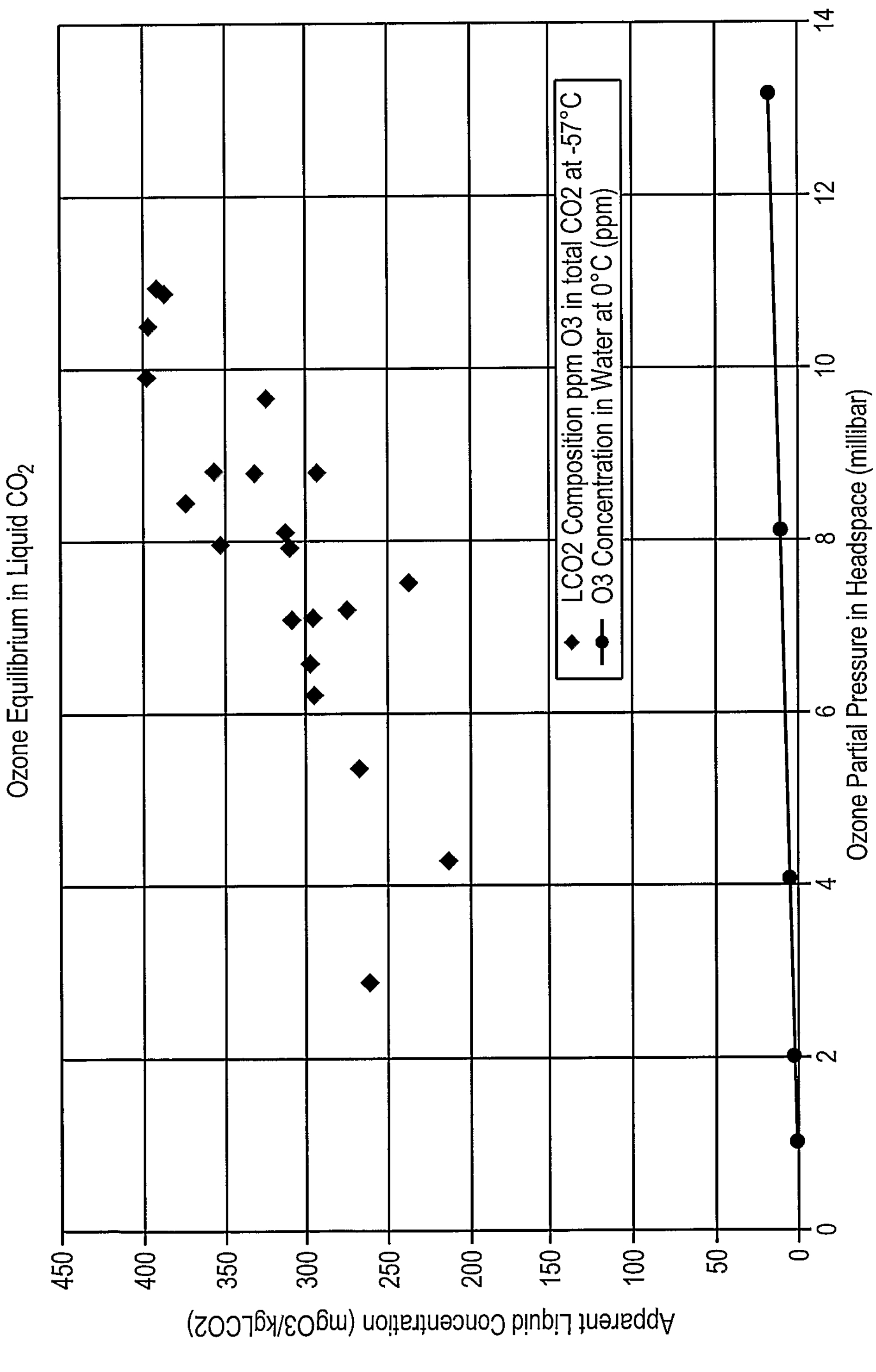
Ozone Equilibrium in Liquid CO_2
Apparent Liquid Concentration (mgO3/kgLCO2)
Ozone Partial Pressure in Headspace (millibar)
LCO2 Composition ppm O3 in total CO2 at -57°C
O3 Concentration in Water at 0°C (ppm)
0 50 100 150 200 250 300 350 400 450
0 2 4 6 8 10 12 14

FIG. 6