

FIG. 14 is an elevation of an apparatus for continuous processing of articles;

FIG. 15 is a plan view of a continuous processing apparatus;

FIG. 16 is a diagrammatic illustration of another apparatus useful in practicing the invention;

FIG. 17 is a plan view of the apparatus of FIG. 16;

FIG. 18 is a diagrammatic plan view of a processing apparatus for practicing the invention as a part of a continuous operation; and

FIG. 19 is a fragmentary elevation taken along line 19—19 of FIG. 18.

As a part of the following description of the invention of the invention, there will be given specific information regarding a preferred mode of performing the process. This specific information represents details of a method which has been found to be effective. However, several variations in the specific steps are discussed herein and others will be apparent.

In FIG. 1 an article 20 is shown in a chamber 21 in which an environment is established for promoting sterilization of the article. The article 20 may be a foodstuff which is sterilized in order to permit storage for an extended period without spoilage, or may be a product which should be utilized in a sterile state.

Chamber 21 is formed by a cylindrical wall member 22, as of glass, with upper and lower plates 23, 24, respectively, sealed to the ends of the cylinder by gaskets 25. The end plates 23, 24, are secured together by tie rod 27 and nut 28. In practice, several tie rods are used, although only one is illustrated in the drawing for simplicity. End plates 23 and 24 are conductive and are connected with an electrical source 30 which establishes an electric field within the chamber 21. The plates are preferably of metal as is tie rod 27. A sleeve 31, of insulating material, surrounds tie rod 27 where it passes through plate 23, electrically isolating the plates. The interior of chamber 21 is connected through a passage 24a in plate 24 with a vacuum pump 33, through a valve 34 with a source 35 of additive gas and through valve 36 with an air inlet.

Inside chamber 21 a platform 40 is mounted on legs 41 above passage 24a. Article 20 is supported above platform 40 on an openwork frame 42, of insulating material, which holds the article without masking the contents.

Briefly, the sterilization operation is carried on in the following manner. Chamber 21 is evacuated by operation of vacuum pump 33 and then a plasma is formed within the chamber by establishing an electric field between plates 23, 24, which serve as electrodes. The term plasma as used herein means an environment in which the molecules are ionized. The ionized molecules of the atmosphere penetrate the package of article 20 and reach any living organisms therein.

If alternating energy is present in the treatment zone within chamber 21 at an appropriate frequency, as in the field exciting the plasma, or in an auxiliary field, electrical currents are induced in the nucleus of the organism cells, and more particularly in the DNA molecule thereof, which alter or destroy the DNA molecule and prevent reproduction of the cell. With the capability of cell reproduction lost, the organism dies. The cells of different organisms respond to currents at different frequencies, permitting selective treatment. The ionized environment provides coupling between the field and the organism.

Ionized (charged) particles of the plasma have sufficient energy to penetrate the protein shell of the organism cells. When the charged particles strike the DNA molecules, cross linkages in the molecule are broken or modified, impairing the reproductive capacity of the organism. The ionized particles are equivalent to the secondary electrons and ions generated by high intensity electron beam sterilization, but are excited directly at a low energy level.

The constituents of the plasma (primarily oxygen and nitrogen with an air environment) react chemically with the organism. Under ordinary conditions, the reactions occur slowly and may have no adverse effect on the organism. With a plasma in which the molecules have an elevated energy level, the reactions are greatly speeded up.

It is important to the process that charged particles of the plasma reach the cells of the organism. Several gases have molecules with greater mobility or a higher molecular coupling coefficient than the constituents of air. Such an additive gas may be introduced into chamber 21 by opening valve 34 after the chamber has been evacuated.

The plasma within chamber 21 may be established by different types of electrical excitation. The spark gap power supply of FIG. 2 has input terminals 45 connected with a suitable electrical source, as 120 volt, 60 cycle AC. A step-up auto transformer 46 is connected with a tuned circuit including capacitor 47 and inductor 48, which is the primary winding of an output transformer 49. Vibrator switch contacts 50, connected in series with inductor 48, are magnetically actuated to close and open 120 times per second by coil 51, connected through on-off switch 52 across the line. When the contact is closed, current flows from capacitor 47 and transformer 46 to inductor 48 and oscillation occurs at a rate determined by the values of capacitor 47 and inductor 48. The output, a burst of oscillatory energy recurring at a 120 cycle rate, is connected from secondary winding 53 to the electrode plates 23 and 24 of chamber 21.

In FIG. 3 a simple 60 cycle AC power supply is shown having an adjustable auto transformer 55 with a primary winding 56 connected through input leads 57 and an on-off switch 58 with a 60-cycle source. The output of the autotransformer is derived from secondary winding 59 through an adjustable tap 60 and is connected with a primary winding 61 of a step-up transformer 62. The secondary winding 63 of the step-up transformer is in turn connected with electrode plates 23 and 24.

A third power supply is illustrated in FIG. 4. A variable frequency driving signal from generator 65 is connected with a higher voltage amplifier 66. A voltage control 67 permits the operator to set the operating conditions of the amplifier and thus the output voltage. In a representative embodiment, the variable frequency generator has a range of 100Hz to 100KHz and the peak voltage output of the amplifier is of the order of 50,000 volts.

The selection of one or a combination of the exciting signals depends on the nature of the organism to be treated. Specific examples will be given below.

FIGS. 5 and 6 are curves illustrating the nature of the ionizing voltage and current, respectively, with the spark gap power supply of FIG. 2. The ionizing voltage comprises a series of energy bursts at a 120 cycle rate (determined by the closure of contacts 50) with each

burst containing several cycles of energy at a fundamental frequency determined by the inductance 48 and capacitance 47 of the tuned circuit. In this case the oscillatory frequency is of the order of 1 kilohertz, but has an irregular pulse shape which results in the presence of many harmonics. Accordingly, the exciting energy covers a wide range of frequencies. The wave-shapes occurring on alternate half-cycles of the 60-cycle energizing voltage are similar in shape but opposite in polarity. The current curve of FIG. 6 shows the major portion of the current flows during the initial half cycle of the energizing voltage establishing the plasma within the chamber. With a chamber having 15 inches spacing between the upper and lower electrodes 23, 24 and a diameter of 10 inches, the voltage waveform of FIG. 5 has a peak-to-peak value of 70 kilovolts and the current waveform has a peak-to-peak value of 0.14 ampere. This gives a peak power rating of the order of 10 KVA. The duty cycle is of the order of 10 per cent, however, so that the average power required is low as compared with the peak power.

The effectiveness of the plasma discharge in killing microorganisms is illustrated in FIG. 7 where the per cent kill is plotted logarithmically as a function of the dosage factor. The dosage factor is a composite term which combines the time duration and the intensity of the plasma discharge. The curve shows that for a particular microorganism a dosage factor can be selected which will reliably effect a 100 per cent kill. Different dosage factors may be necessary for different types of organisms. The plasma discharge does not appreciably raise the temperature of the matter treated. However, it has been found that efficiency of the process may be improved by operating at an elevated temperature, but below a temperature at which the matter is modified.

It is my theory that several different factors contribute to the sterilizing effect of the processes described herein. First, the alternating fields coupled through the plasma to the organisms set up disruptive currents which alter the DNA molecules to an extent sufficient to prevent reproduction. I believe these currents are complex functions of electron and ion mobilities and concentrations, in the nature of currents in semiconductors. Complex resonant current conditions have been observed in effecting a complete kill. Although circulating electric currents are believed to be a major factor in disrupting the ability of the DNA molecule to control reproduction of the organism, more complex effects also take place, including electron spin resonance and multiple nuclear magnetic resonance, both of which modify the characteristics of the organism. All these effects are believed to be caused, at least to some extent, by subjection of the organisms to an alternating field, electric, magnetic or electromagnetic, at an appropriate frequency or combination of frequencies. The plasma improves the energy transfer from the fields to the organism.

Second, charged particles of the plasma penetrate the microorganisms and, upon striking a cell, give up energy which breaks bonds within the DNA molecules. If, for example, the phosphate-sugar bond is broken, reproduction is inhibited. The effectiveness of this aspect of the process depends on the relation between the mean-free path of the particles (i.e., the distance they travel before striking an organism or another particle) and the organism size. Preferably, the mean-free path is greater than 5 or even 10 organism diameters. This

characteristic is related to the level of vacuum in which the plasma is established, the mean-free path being an inverse function of the pressure. The significance of this characteristic is illustrated in FIG. 8 where charged particles follow paths 75, 76 and 77 through organism cells 78. The charged particles which follow paths 76 and 77 are shown striking cells 78a and 78b while the particle which follows path 75 passes between the organism cells and strikes another particle at X. If the particle population is large (high pressure), most of the charged particles strike other particles when they have traveled a distance which is short with respect to the organism size, and very few particles will strike an organism. Accordingly, it is important that the pressure be reduced to increase the mean-free path of the charged particles.

Third, a chemical reaction between the plasma constituents and the organism destroys the organism. This is analogous to the alkylation of gas sterilization, but much faster. The energy of the particles resulting from the excited condition of the plasma speeds up the chemical reaction so that even oxygen or nitrogen from the atmosphere are effective sterilizing agents. More common sterilizers, as ethylene oxide, may be used, with a further time saving.

It is estimated that approximately 50 per cent of the energy of the charged particles is dissipated in penetrating the cell while the other 50 per cent is utilized in destroying the capacity of the organism to reproduce. The diagrammatic representation of a typical cell in FIG. 9 shows that the nucleus 80 is surrounded by a shell 81 which in turn is surrounded by a protein layer 82. The gaseous constituents of atmosphere, principally oxygen and nitrogen, are not particularly efficient in penetrating the protein layer and shell of an organism. The treatment can be improved by utilizing a gas which is more effective in this respect. Examples are ethylene oxide and  $\text{CCl}_2\text{F}_2$ , one of the gases sold as a refrigerant under the trademark FREON. When ethylene oxide is used, the chemical action of the process is also enhanced.

A catalyst (as cesium) may be used to modify the characteristics of the plasma, permitting an increase of power without the use of excessive voltages. Above 30KV some shielding may be desirable to limit the radiation of soft X-rays. Such shielding is much simpler than that used for present high intensity radiation.

The DNA molecule has a spiral chain link configuration illustrated in FIG. 9 with a variety of chemical links between the longitudinal members 84, illustrated by the dashed lines 83. This cross-linked construction permits the establishment of a variety of circulatory currents in different paths, indicated by the arrows. The various paths have different resonant characteristics and permit selective modification in the DNA molecule by subjecting it to excitation at a frequency or frequencies which establish currents in some paths but not in others.

A control over the sterilization process is commonly made by including in one or more of the articles treated a spore strip of an organism which is particularly difficult to kill. One such organism is bacillus subtilis var. globigii. In a typical test, a strip having  $10^6$  organisms thereon is placed in the article to be treated. Following treatment, the strip, or a sample from the strip, is incubated to determine whether the organisms are capable of multiplication.

With the apparatus of FIG. 1 and the power supplies of FIGS. 2 and 3, consistent kills have been achieved using the following procedure:

1. Evacuate chamber 21 to a pressure of 300 microns;
  2. Energize with the spark gap power supply of FIG. 2 for a period of 3 minutes;
  3. Open air inlet valve 36 to return the chamber to atmospheric pressure;
  4. Evacuate again to a pressure of 300 microns;
  5. Add a gas (as  $\text{CCl}_2\text{F}_2$ ) to a pressure of 5,000 microns, permitting thorough penetration of the article by the gas;
  6. Evacuate to 1,500 microns;
  7. Energize from the spark gap source for 3 minutes.
- At this point in the treatment, consistent 99 per cent kills have been achieved. It is believed that losses in the spark gap power supply have reduced the treatment efficiency so that the kill is not 100 per cent effective. The following additional steps insure complete kill:
8. Admit air to atmospheric pressure;
  9. Evacuate to 500 microns;
  10. Energize from the power supply source of FIG. 3 with a voltage of 1,500 volts, RMS (and a current of the order of 0.5-0.9 amps);
  11. Admit air to atmospheric pressure;
  12. Evacuate to 500 microns;
  13. Add  $\text{CCl}_2\text{F}_2$  to 5,000 microns;
  14. Evacuate to 1,500 microns;
  15. Apply 3 minutes power from the power supply of FIG. 3.

With the power supply of FIG. 4, the following procedure has been found effective for the vacillus subtilis:

1. Evacuate to 300 microns;
2. Energize for 3 minutes, varying the frequency from 2,000 $\text{H}_z$  to 20,000 $\text{H}_z$  at each of three peak-to-peak voltage settings, 120KV, 160KV and 200KV;
3. Evacuate to 300 microns;
4. Add  $\text{CCl}_2\text{F}_2$  to 4,500 microns;
5. Energize for 2 minutes, 20 seconds, as in (2) above.

Salmonella paraphi have been killed with this treatment:

1. Evacuate to 300 microns;
2. Energize for 5 minutes (100 seconds each)
  - 2,000 $\text{H}_z$  at 120KV
  - 5,000  $\text{H}_z$  at 150KV
  - 10,000 $\text{H}_z$  at 170KV;
3. Evacuate to 300 microns;
4. Add  $\text{CCl}_2\text{F}_2$  to 4,500 microns;
5. Evacuate to 500 microns;
6. Energize for 5 minutes (100 seconds each)
  - 2,000 $\text{H}_z$  at 140KV
  - 5,000 $\text{H}_z$  at 150KV
  - 10,000 $\text{H}_z$  at 190KV;
7. Add  $\text{CCl}_2\text{F}_2$  at 1,000 microns;
8. Energize for 6 minutes 40 seconds (100 seconds each)
  - 2,000 $\text{H}_z$  at 300KV
  - 5,000 $\text{H}_z$  at 220KV
  - 10,000 $\text{H}_z$  at 200KV
  - 20,000 $\text{H}_z$  at 160KV.

Sterilization has been effected within a tin coated steel can. An ordinary can of tuna fish from a grocer's shelf was opened and a globigii spore strip placed in-

side. The cover of the can was resealed with solder. The can was then subjected to the following treatment:

1. Evacuate to 500 microns;
2. Energize for 3 minutes at 16,000 $\text{H}_z$ , 40KV;
3. Evacuated to 500 microns;
4. Energized for 3 minutes at 1,400 $\text{H}_z$  and 55KV.

The spore strip was incubated. At the end of 12 hours, there was an indication that the globigii had grown. The rate of growth then diminished and at the end of 72 hours no globigii remained alive. A globigii control strip incubated with the test strip showed continued growth throughout the 72 hour period.

An apparatus is illustrated in FIG. 11 for exciting a plasma with different types of energy. An elongated cylindrical body 90, as of glass, defines the space within which the plasma may be formed, and has transverse extensions 91, 92 and 93, 94 forming crosses therewith adjacent either end. The ends of the cylindrical body and of each of the cross extensions are closed by plates 95 through which connections may be made to the interior of the body. Extending outwardly from the cover members for extensions 91 and 92 are supporting tubes 96 for electrode plates as 97, 98, 99 and 100, within the extensions. In the specific structure of the drawing, electrode 97 cooperates with electrode 100 while electrode 98 cooperates with electrode 99, so that electric fields are established therebetween when the electrodes are energized as by power supplies of the character shown in FIGS. 2 and 3. Focusing electrodes 103 may be associated with each of the field establishing electrodes. Adjacent electrode assemblies (i.e., electrode 97 and its associated focusing electrode 103) are isolated from the adjacent electrode assemblies by shields 104. The article 105 to be treated is supported in the treatment zone on a platform 106 at the intersection of the main body and the transverse extensions 91, 92.

The apparatus is evacuated by a pump 108 connected through valve 109 with extension 94. Test probes as Langmuir or Faraday probes may be introduced into the interior of the chamber through axial extensions 110, at either end. Gauges, as for pressure level and temperature, may be introduced into extension 93 at 111.

Magnetic coils 112, 113, 114, 115 placed around the cylindrical housing and extensions 91, 92, surrounding the position of the article being sterilized, provide excitation the excitation of the plasma magnetically at one or several frequencies. Furthermore, suitable energization of the coils with direct current sets up a "magnetic bottle" or "magnetic well" effect which concentrates and contains the plasma.

FIG. 12 illustrates schematically another apparatus for electrically stimulating a plasma discharge at a plurality of frequencies. Here a cylindrical chamber 120 has therein three sets of opposed electrodes 121, 121', 122, 122', and 123, 123'. Each pair of electrode is connected with the corresponding signal generator, 121a, 122a and 123a. The signal generators may be operated at the same or at different frequencies depending on the nature of the organism (not shown) to be treated. Shield members (shown diagrammatically at 124) are interposed between adjacent electrodes.

FIG. 13 illustrates a specific structural arrangement in which adjacent electrodes 127 are isolated by portions 128 of the wall of chamber 129 which extend inwardly beyond the location of the electrodes.

In FIG. 14 an apparatus is shown for the continuous processing of articles 135 carried through a treatment zone on conveyor belt 136. A plasma is formed in the atmosphere and the organisms within the articles are excited by electromagnetic radiation from a horn radiator 137 connected through waveguide 138 with a suitable source, as a signal generator, not shown. A second horn radiator 139 is connected with another source of power through waveguide 140 and is located below conveyor 136, directing radiated energy upwardly toward articles 135. If treatment at additional frequencies is desirable, further radiators may be placed at other points along the length of conveyor 136. Alternatively, as shown in FIG. 15, the articles on conveyor 136 may also be subjected to a magnetic field (or a plurality of them) established by currents flowing through coils 142. The pole pieces 143 associated with each coil serve to concentrate the flux in the path of the articles.

FIGS. 16 and 17 illustrate an apparatus for applying a unidirectional magnetic orienting field to the matter being treated. The chamber 150 defining the treatment zone is again provided by cylinder 151 closed by conductive end plates 152, 153. Details of the connections with the electrical circuit, the vacuum pump and the source of additive gas are not shown, but may be generally the same as that illustrated in FIG. 1. Article 154, to be treated, is located within chamber 150. A magnetic structure 155 has a pair of tapered poles 156 diametrically located on either side of chamber 150 and intermediate the ends thereof. A pair of coils 157 on core 155 are energized by direct current to establish a high intensity unidirectional magnetic field within the chamber 150. The unidirectional field may be of the order of at least one kilogauss and may be as high as 20 kilogauss, depending on the nature of the organisms treated. The intense magnetic field serves to orient the molecular structure of the organism to be treated in a plane generally at right angles to the axis of the plasma. In addition to the electrical field established between the plates 152 and 153, the plasma may be excited magnetically by coils 160, 161 wound around the cylinder 151. The alternating plasma fields from coils 160, 161 are also at right angles to the unidirectional magnetic orienting field. The orienting field coils need be energized only when the plasma is excited.

The establishment of a uniform orientation for the structure of the organism improves the frequency selective characteristics of the process so that the organisms may be treated with a lesser dosage factor than is required when they are not oriented. Coils 163, 164, 165 and 166 extend generally the length of cylinder 151 and have their axes parallel therewith. They are energized with direct current, the polarity of diametrically opposed coils being the same, as indicated in FIG. 17. These coils focus and concentrate the plasma in the center of chamber 150, for maximum effectiveness in treatment of organisms located at the center of the treatment zone.

In FIGS. 18 and 19 an apparatus is shown for performing the process as a part of a continuous production line. A plurality of enclosures 170 are mounted on a conveyor 171 for travel in a closed loop 172. Each container has a top 173 and a bottom 174 which may be opened by a suitable mechanism (not shown); and is attached by a flexible connector 176 with a distributor 177 mounted at the center of the conveyor loop.

Each container 170 is airtight when closed and may be evacuated. Energizing electrodes and coils, as those illustrated in FIGS. 1, 11 and 16, and otherwise disclosed herein, are provided for the container, though they are not illustrated in detail. Flexible connector 176 includes multiple passages and electrical conductors for establishment of a desired environment in the treatment zone of the associated containers 170 and for the excitation of a plasma therein.

Distributor 177 has a fixed portion 177a to which are connected suitable conduits and electrical cables 178, 179, 180 from vacuum pump 182, gas source 183 and electrical source 184, respectively. The upper portion 177b of rotary distributor 177 turns with respect to the fixed base 177a in synchronism with movement of containers 170 around loop 172, and suitable porting and switching (not shown) controls the connection of each container with the vacuum pump, gas or electrical source at proper times.

At loading station 186 the cover 173 of container 170 is opened and an article 187 deposited therein as from a belt conveyor 188. Cover 173 then closes and the sterilization process commences. After the container 170 has traveled around the conveyor loop 172, container bottom 174 is opened at a discharge station 190 and article 187 is deposited on a conveyor belt 191 which moves it on for further handling.

It has been found that the sterilization process may sometimes be more efficient at an elevated temperature. Accordingly, a portion of conveyor loop 172 may extend through a heating tunnel, the entrance and exit of which are indicated by broken lines 192, 193.

While I have shown and described certain embodiments of my invention, it is to be understood that it is capable of many modifications. Changes, therefore, in the construction and arrangement may be made without departing from the spirit and scope of the invention as without in the appended claims.

I claim:

1. A method of modifying the reproductive mechanism of an organism which comprises establishing a rarefied atmosphere, establishing a low energy plasma by exciting said rarefied atmosphere with alternating energy comprising a plurality of discrete frequencies, said energy including electromagnetic radiation and alternating magnetic fields at different frequencies, and exposing the organism to the plasma.

2. A method of modifying the reproductive mechanism of an organism which comprises establishing a rarefied atmosphere, establishing a low energy plasma by exciting the rarefied atmosphere with an electric field including a wide range of frequencies, having a basic frequency of the order of 10 kilohertz, established periodically at a repetition rate of 120 Hertz and exposing the organism to the plasma.

3. The method of claim 2 in which the ratio of average power to peak power in establishing said field is of the order of 0.1.

4. A method of modifying the reproductive mechanism of an organism which comprises establishing a rarefied atmosphere, establishing a low energy plasma having an axis by excitation of said rarefied atmosphere and aligning the molecular structure of the organism with respect to the plasma by the application of a unidirectional magnetic field at right angles to the plasma axis.

5. A method of modifying the reproductive mechanism of an organism which comprises establishing a rarefied atmosphere, establishing a low energy plasma by exciting said rarefied atmosphere with alternating energy covering a wide range of frequencies applied sequentially and exposing the organism to the plasma.

6. A method of modifying the reproductive mechanism of an organism which comprises establishing a rarefied atmosphere, establishing a low energy plasma by exciting said rarefied atmosphere with alternating energy at a first frequency pulsed at a lower frequency, said pulsation at said lower frequency creating a wide range of excitation frequencies, and exposing the organism to the plasma.

7. A method of modifying the reproductive mechanism of an organism which comprises establishing a rarefied atmosphere, establishing a low energy plasma by exciting said rarefied atmosphere with alternating energy comprising a plurality of discrete frequencies applied sequentially and exposing the organism to the plasma.

8. A method of modifying the reproductive mechanism of an organism which comprises establishing a rarefied atmosphere in a medium selected from a group comprising ethylene oxide, and a paraffin hydrocarbon with one or more fluorine atoms, establishing a low energy plasma by excitation of said rarefied atmosphere and exposing the organism to the plasma.

9. An apparatus for creating a low energy plasma, comprising;

means defining a treatment zone;  
field establishing means operatively associated with said zone and including a plurality of pairs of spaced electrodes;

means providing an electric shield between adjacent electrodes;

a plurality of sources of alternating energy at different frequencies connected with different pairs of said electrodes to create a plasma in said zone; and means within said zone for supporting matter to be subjected to the plasma.

10. An apparatus for sterilizing matter, comprising: a plurality of chambers traveling in a closed path between a loading and a discharging station, each chamber having field establishing means operably associated therewith;

means operable during travel of each chamber from the loading station to the discharge station for establishing a treatment zone therein, including means for maintaining a rarefied atmosphere in the chamber and a source of electrical energy connected with said field establishing means to create a low energy plasma in the chamber;

means within each chamber for supporting matter to be subjected to the plasma; and

conveyor means for delivering matter to be sterilized to said loading zone and for removing sterilized matter from the discharge zone.

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