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METHOD AND APPARATUS FOR REACTING IONIZED GAS WITH A NON-GASEOUS SUBSTANCE

Filed Aug. 19, 1968 2 Sheets-Sheet 1

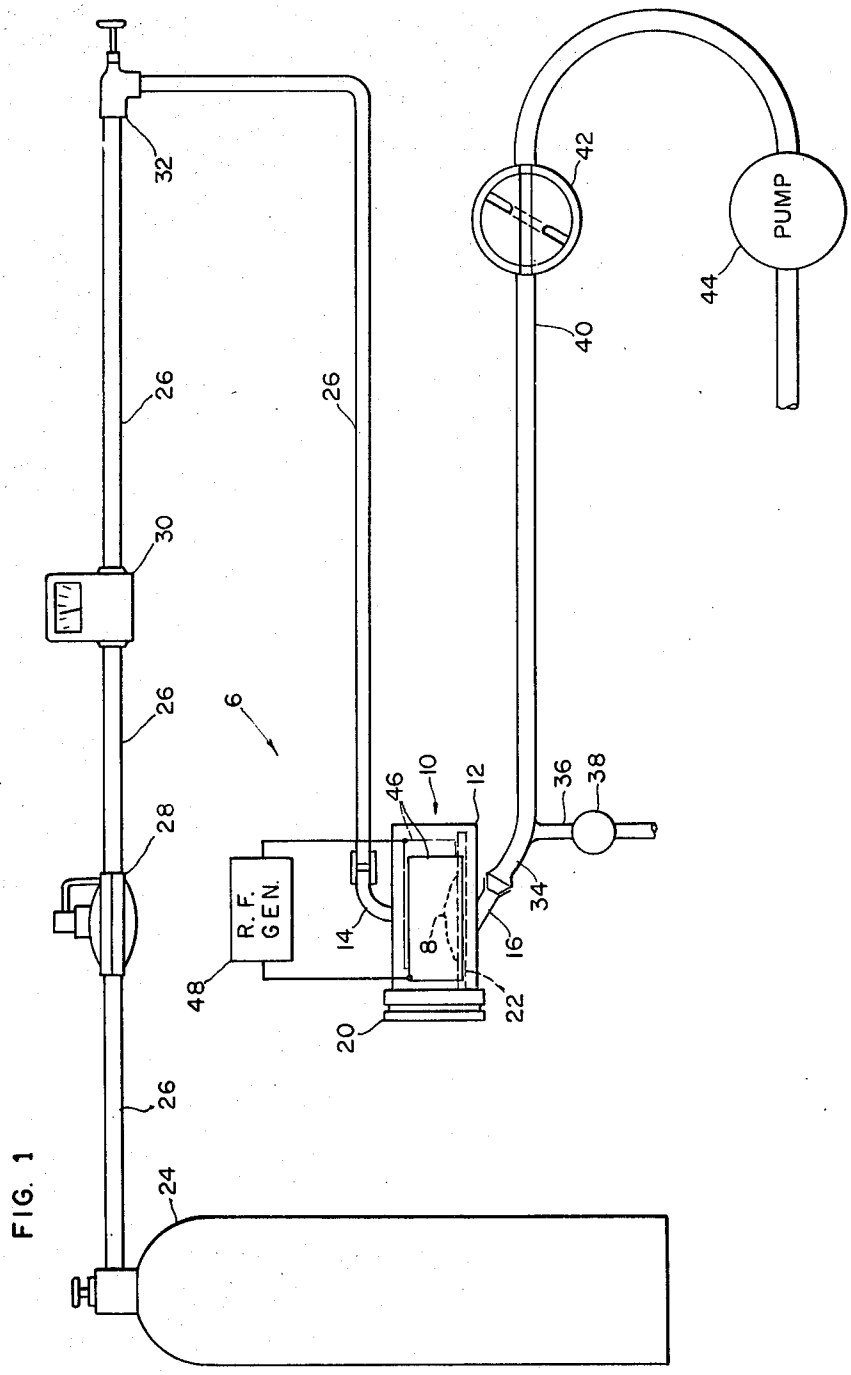


FIG. 1

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FIG. 2

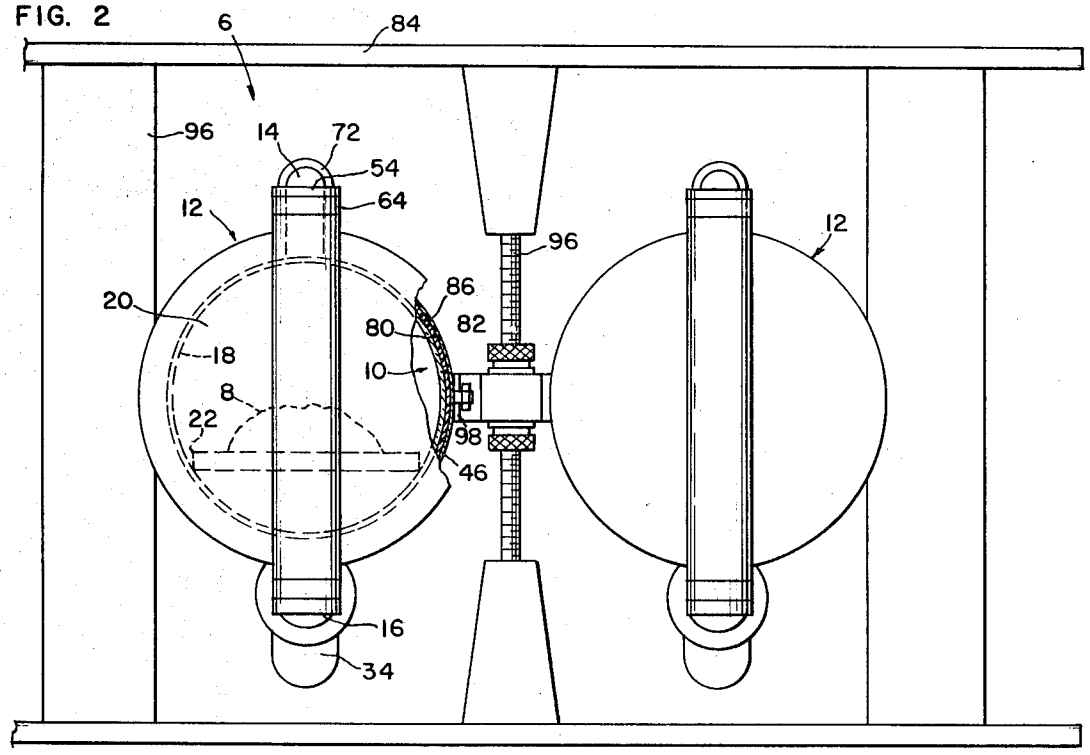
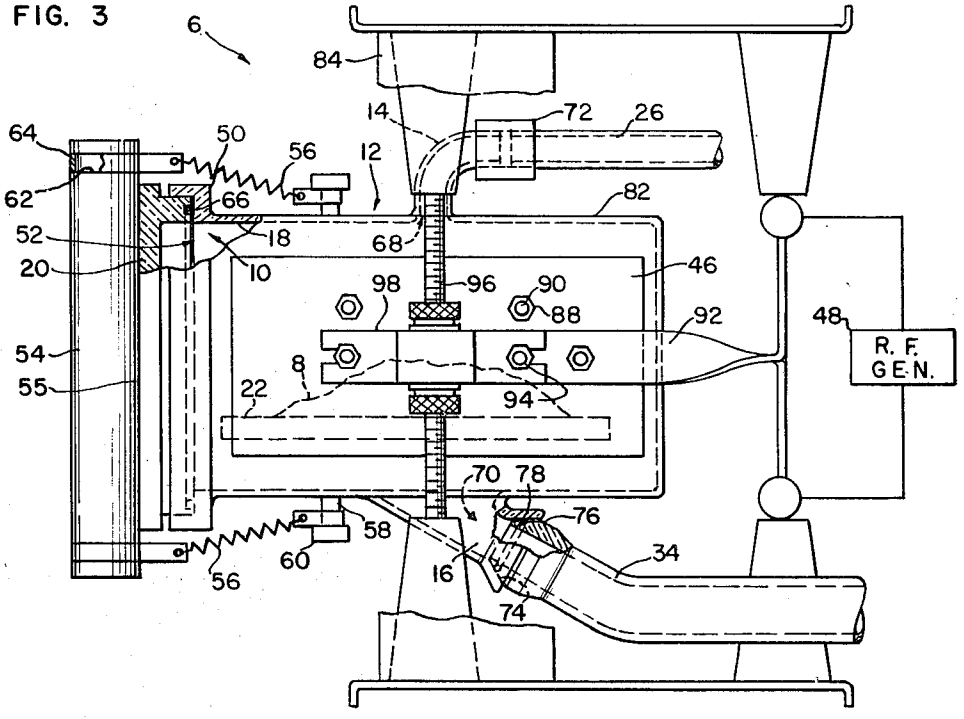


FIG. 3



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METHOD AND APPARATUS FOR REACTING IONIZED GAS WITH A NON-GASEOUS SUBSTANCE

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Int. Cl. B01k 1/00
U.S. Cl. 204—312 4 Claims

ABSTRACT OF THE DISCLOSURE

A method for ionizing gas and reacting it with a non-gaseous substance in which the gas is ionized by an electrical radio frequency (RF) field generated in the space surrounding the substance. The apparatus comprises a container defining a reaction chamber and a pair of electrodes coupled with an RF generator and placed closely adjacent an exterior surface of the container. The container includes an intake conduit and an exhaust conduit which is preferably diammetrically opposite the former. An access opening to the chamber is closed and sealed from the exterior of the container by a cover biased toward the portion of the container defining the opening.

RELATED APPLICATIONS

This application is related to the commonly owned copending patent application bearing Ser. No. 753,682, filed Aug. 19, 1968, for Plasma Generating Apparatus, now U.S. Pat. No. 3,573,192 patented as set forth in the body of this specification.

BACKGROUND OF THE INVENTION

This invention relates to plasma generating apparatus and method and more particularly to a plasma generating apparatus and method in which gas is ionized in the space surrounding a non-gaseous substance with which the plasma, i.e. the ionized gas, is to be reacted.

Low temperature, ionized gas, or cold plasma, has become widely employed for reaction with non-gaseous substances for processing as well as analytical purposes. For example, to enumerate but two of many such purposes, surface characteristics of certain plastics may be altered by subjecting the plastics to the plasma; organic substances such as human, animal, or plant tissue may be oxidized, or ashed, to obtain an inorganic residue without destruction or loss of the residue from high temperatures and/or volitization. The substance to be treated, that is the specimen or sample, is placed in a reaction chamber for a sufficient length of time to achieve the desired changes in its characteristics or to obtain a complete oxidation of the organic ingredients in the sample.

According to the prior art, gas, such as oxygen, for example, is passed into an ionization chamber between induction coils which subject the chamber and the gas to an electrical radio frequency (RF) field and thereby energize and ionize the gas. From the ionization chamber, the gas is passed to a reaction chamber containing the sample and the two are reacted in the chamber. Gas is continuously introduced into the chambers and withdrawn or exhausted therefrom at like rates.

Although a plasma breakdown, or return of the plasma to its molecular state, occurs as soon as the plasma leaves the RF field, it has been thought that the placement of the sample directly into the RF field results in excess reaction between the plasma and the sample and in a rise of the temperature of the sample beyond tolerable limits. The plasma breakdown, however, results in appreciable energy losses which were heretofore thought to be unavoidable.

It is frequently desired to place a plurality, often as many as five, reaction chambers in a single apparatus to provide for flexibility of the apparatus and the increase of its capacity. Some prior art plasma generating machines employ reaction chambers from which the ionization chambers extend sideways. The ionization chambers are disposed at about the longitudinal center of the reaction chambers to evenly distribute plasma throughout the reaction chamber; the ionization chambers are substantially smaller than the reaction chambers; and they are arranged in an aligned side-by-side relation to permit the placement of single induction coils or capacitive rings around all chambers.

Although economical to construct, the single coils or rings result in non-uniform RF fields in the various ionization chambers since the intensity of the field varies with the position of the particular chamber with respect to the coil. For example, chambers adjacent the loop of the coil are subjected to an RF field of substantially greater intensity than that imposed on intermediate chambers. At the coil loop the effective length of the coil is substantially greater than at the intermediate chambers because in the case of the former, the coil is wrapped around the chambers, while at the latter the coil extends merely parallel thereto. The degree of ionization of the gas, therefore, varies from ionization chamber to ionization chamber and results in non-uniform reactions between the plasma and the specimens in the several reaction chambers of the apparatus.

In addition, removal of one of the chambers from the apparatus causes serious disturbances in the plasma generation in the other ionization chambers. It has not been possible to determine the exact causes for this phenomena.

Lastly, the relatively small size of the ionization chambers and the resulting proximity of the gas inlet causes a partial ionization of gas in the supply conduits to the chamber. Unless inert materials, such as glass, are used for such conduits ionized gas damages and destroys such conduits and their fittings. To avoid this problem capillary entry conduits into the ionization chambers must be provided. Although capillary entry conduits reduce or eliminate damage to the supply conduits, they cause large pressure drops and generally a reduction in the desired optimal gas flow rate through the ionization chamber and vicinity of the specimen being treated.

SUMMARY OF THE INVENTION

The present invention provides a method and apparatus for reacting ionized gas with a non-gaseous specimen or sample in a sealed reaction chamber defined by the container of the apparatus. Briefly, the method of the invention comprises the steps of placing the specimen in the chamber, passing the gas through the chamber at a predetermined pressure, and applying a radio frequency (RF) electrical field over that portion of the chamber in which the sample is positioned. Gas surrounding the substance is thereby ionized to minimize energy losses from a breakdown of the plasma before it can reach the sample.

The apparatus of the invention comprises a support structure and a container mounted thereon. The container defines a reaction chamber and includes a gas inlet, a gas outlet, and an access opening. A cover closes the access opening and seals the chamber from the exterior of the container. A pair of opposing electrodes are positioned on the exterior of the container at a location permitting the placement of the specimen to be treated between the electrodes. Means for electrically connecting the electrode with a radio frequency (RF) generator enables the energization of the electrodes by the generator so as to subject a portion of the chamber to an RF field which ionizes the gas being circulated through the chamber.

In the presently preferred embodiment of the invention, the electrodes comprise electrode plates spaced from the container by an insulating material in contact with both the container and the plates. The electrode plates extend over substantially the full axial length of the container to permit the placement of even the largest samples that can be received in the reaction chamber in the RF field. The plates are constructed to frictionally engage the container and permit its slideable axial movement between the electrodes. An intake and an outlet conduit extend from the container and have free ends facing in an axial direction so that the slideable insertion of the container between the plates automatically couples the conduits with supply and exhaust conduits of the apparatus.

It has been found that even though the samples are surrounded by the RF field, no appreciable temperature rise above that encountered in prior art plasma generating and reacting apparatus is experienced. Thus, a typical apparatus may operate under a gas pressure in the reaction chamber of a few millimeters mercury and be subjected to an RF field having a frequency of about 13.6 megacycles as in prior art plasma generating machines; yet, the temperature of the sample being reacted does not rise above about 130° C. Consequently, damage to the sample from excessive temperatures or volitization is avoided while permitting a maximum utilization of the available RF energy and significantly increasing the reaction rate between the sample and the plasma.

In addition, the plasma does not travel back into the intake conduits even if that conduit is relatively large and non-capilary. The heretofore experienced damage to supply conduit components is eliminated. Relatively high gas flow rates can thereby be obtained without excessive speeds of the gas in a capillary section of the intake conduit as was common in prior art plasma generating machines. Undesirable disturbances in the reaction chamber from high velocity gases, unless first introduced into a separate, i.e. an ionization, chamber, are eliminated.

In addition, the arrangement of the electrodes and their position relative to the container permit that the container be suspended from the electrodes. The container is much more accessible, and can be removed for replacement or repair, for example, as more fully set forth in the above referred to copending patent application.

BRIEF DESCRIPTION OF THE DRAWING

FIG. 1 is a schematic representation of an apparatus for reacting ionized gas with a non-gaseous substance constructed in accordance with the present invention;

FIG. 2 is a fragmentary, side elevational view of the apparatus and shows the container defining the reaction chamber in greater detail; and

FIG. 3 is a fragmentary, side elevational view, with parts broken away, of the apparatus shown in FIG. 2.

DESCRIPTION OF THE PREFERRED EMBODIMENT

FIG. 1 schematically illustrates an apparatus for generating ionized gas or plasma in a radio frequency (RF) field. The plasma is reacted with a non-gaseous substance or sample **8**, such as ground human tissue for example, in a reaction chamber **10** defined by a container **12**. The container includes an intake opening **68** from which an intake conduit **14** extends, and an outlet opening **70** from which an outlet conduit **16** extends. One end of the container is defined by an access opening **18** closed by a cover **20** to enable the insertion and withdrawal of the sample which is preferably disposed on a tray or boat **22**.

A source of gas, such as a bottle **24**, that may contain molecular oxygen gas, for example, is connected with the intake conduit through supply conduit **26**. The supply conduit preferably includes a pressure regulator **28**, a flow meter **30**, and a needle valve **32** for conventional regulation and control of the flow rate and gas pressure at the intake conduit. The outlet conduit connects to exhaust conduit **34** which branches into a bleeder line **36**, closed by bleed valve **38**, and a suction line **40** that may include a two-stage valve **42** and a vacuum pump **44** exhausting to the atmosphere.

A pair of opposing electrode plates **46** are disposed on the exterior of container **12**, as more fully set forth below, and each plate is connected with an electrical RF generator **48**. An electrical RF field between the plates encompasses the space of chamber **10** in which sample **8** is disposed.

In operation, boat **22** carrying sample **8** is inserted into reaction chamber **10** through access opening **18**, the opening is closed with cover **20**, bleed valve **38** is closed, and vacuum pump **44** is actuated to reduce the pressure in the reaction chamber to the desired level, say one or two millimeters mercury. A fast pump down is obtained by setting valve **42** to its full open position. A slower pump down rate is obtained by setting the two-stage valve to its position shown in FIG. 1. After the desired vacuum in the reaction chamber has been attained and with pressure regulator **28** set for the desired pressure, i.e. 1 to 2 millimeter mercury, needle valve **32** is opened until flow meter **30** indicates the desired flow rate while the operation of the vacuum pump is continued so that oxygen passes through the intake of container **12**, around sample **8**, through outlet conduit **16** and into exhaust conduit **34** to pump **44**. Generator **48** is activated to energize electrode-plates **46** and subject the portion of the reaction chamber between the plates to an RF field which ionizes the oxygen in the chamber. The oxygen plasma now reacts with the human tissues or other specimen forming sample **8** and slowly oxidizes or ashes it at relatively low temperatures to leave a residue not damaged by high temperatures or lost through volatization. At the end of the process pump **44** is de-energized and bleed valve **38** is opened to bring the reaction chamber back to atmospheric pressure.

While the foregoing description has been with reference to low temperature ashing or oxidation, it is understood that the invention herein may be utilized in respect to a variety of other types of reactions employing plasma gases in apparatus of the general type described herein.

In actual practice, RF generators oscillating at about 13 megacycles per second have been found to work out well. The voltage on the plates is, of course, a function of the distance between them and is selected according to well known principles. Similarly, the power output or wattage of the generator is a function of the size as well as the number of electrode plates that are connected with it and is selected accordingly.

FIGS. 2 and 3 show a plasma generating and reacting apparatus **6** which is provided with two adjacent containers **12**. The number of containers can, of course, be varied to increase or decrease the capacity of the apparatus.

The container has a preferably cylindrical cross section, is closed at one end, and includes an annular flange **50** around access opening **18** at the other end of the container. The flange has a flat, annular surface **52** against which cover **20** is biased by an elongate bar **54** engaging a flat face **55** of the cover. Each ends of the bars is engaged by a helical tension spring **56** anchored on a pair of opposite posts **58** which extend from the periphery of the container and are spaced from the annular flange. The free end of each post includes a head **60** to retain the springs on the posts; and bar **54** preferably includes a pair of grooves **62** engaging mounting members **64** for the springs. Bar **54** thereby biases the cover into engagement with annular flange **50**. A cylindrical protrusion **65** on the cover is disposed interiorly of annular flange **50** of the container and positions the cover on the container. A seal ring **66** disposed in an annular groove of the cover seals access opening **18** from the exterior of the container. It will be noted that when reaction chamber **10** is subjected to a vacuum the pressure differential between the chamber and the exterior of the container increases the

force with which the cover is biased against the annular flange.

Inlet opening **68** and outlet opening **70** of the container are preferably diametrically opposed, that is they are opposite and aligned, and both intake conduit **14** and outlet conduit **16** are formed so that their respective free ends face in the direction of the axis of the container and away from cover **20**. The intake conduit has a generally cylindrical configuration throughout its length and its end is disposed in a sleeve-like coupling **72** which is also connected to the free end of supply conduit **26**. The coupling can be constructed of a plastic such as polyvinylchloride (PVC) even though PVC is attacked by certain plasmas, such as oxygen, since as described above, the plasma does not reach back from the reaction chamber to the end of the intake conduit.

Outlet conduit **16** extends away from the container at an angle to the axis of the container and its free end also faces away from cover **20**. The free end of the outlet conduit includes an internal frusto-conical portion **74**, and the end of exhaust conduit **34** has a mating external frusto-conical portion **76** in engagement with portion **74** to provide a connection between the outlet conduit and the exhaust conduit. The conical surfaces are ground for a close fit, and a seal ring **78** is disposed in a groove on conical portion **76** to seal the connection between the two conduits.

It will be noted that when the reaction chamber **10** is vacuumized the pressure differential between the chamber and the exterior of container **12** firmly presses the frusto-conical portions into mutual engagement to provide a positive seal. Similarly, the pressure differential biases the free end of intake conduit **14** into sleeve **72** and provides a positive seal between the two.

Referring to FIGS. 2 and 3, a preferred but not exclusive embodiment of the invention provides the pair of electrode plates **46** disposed on opposing sides of the container **12**; and electrode surfaces **80** facing the container are parallel to an exterior side **82** of the container, i.e. the electrodes are formed to have a circularly arcuate configuration with a radius of curvature somewhat greater than the radius of the exterior side of the container by the distance the plates are spaced from the container. The electrodes are mounted on a support structure **84**, as more fully described in the above referenced co-pending application, that allows for lateral movement of the plates toward and away from the container.

Although the container is constructed of glass, which is normally an insulator, high frequency electrical fields, such as RF fields transform it into a conductor so that direct contact between the container and the electrode plates would result in a short circuit and would damage, i.e. melt, the glass at its contact point with the electrode plates. Consequently a layer 86 of an insulating material is disposed between the electrode plates and the container. This layer serves a double function. First, it provides the desired insulation between the electrode plates and the container; and, second, it enables an equal spacing between the plates and the container without requiring a high precision mounting for the plates. Such an equal spacing is necessary to maintain a uniform RF field throughout the space of reaction chamber **10** between the electrode plates.

It is presently preferred to construct the layer **86** of insulating material in the form of a plastic sheet, such as the plastic known in the industry under the trademark Teflon by the du Pont de Nemours Company of Wilmington, Del., for example. The plastic sheet is secured to the electrode plate by threaded flat head machine screws **88** constructed of an insulating material, extending through apertures (not shown) of the electrode plates, and secured to the plates and the insulating sheet with nuts **90**.

Terminals strips **92** are secured to the sides of electrode plates **46** facing away from container **12** with threaded bolts **84** and extending rearwardly, that is away from cover **20**, for connection to the RF generator **48**. Details of that connection are fully disclosed in the above referenced co-pending patent application.

The electrode plates are mounted to upright posts **96** of the support structure in any convenient manner. It is preferred, however, to mount the electrode plates so that at least one of them is laterally movable toward and away from the container as by employing a leaf spring **98** secured to the electrode plate and mounted to the upright posts. The plates may additionally be adjustable in a vertical direction along posts **96** to position them them in exact alignment with each other. Such a preferred construction is also disclosed in the above referenced patent application and is, therefore, not further treated herein.

To operate apparatus **6** of this invention, container **12** is first placed intermediate the set of electrode plates by moving the container parallel to its axis in between the plates. If the plates are laterally movable, they are constructed so that leaf spring **98** biases one of the electrode plates toward the container and the other electrode plate to frictionally engage the container and mount it to the support structure. The axial movement is continued until the ends of intake conduit exterior frusto-conical portion **76** of exhaust conduit **34**, respectively. It will be noted that the engagement is automatic due to the positioning of the conduits and immediately provides a seal tight engagement between them. Tedious alignment and assembly work with threaded fittings and the like is thus unnecessary.

Bar **54** is now disengaged from cover **20** by sliding it off cover **20** to the left or right, as viewed in FIG. 2, and tray **22** with sample **8** to be treated is inserted into reaction chamber **10**. The tray may have any desired configuration or may be omitted if other suitable means are provided for supporting the sample and positioning it between the electrode plates. Cover **20** is placed over access opening **18** and bar **54** is pulled forward and moved to the center of the cover to engage face **55** of the latter and bias the cover against flange **50** of the container. Reaction chamber **10** is now sealed from the exterior and can be vacuumized. As described above, the decreasing pressure in the reaction chamber increases the seal between container **12** and cover **20**, between intake conduit **14** and intake coupling **72**, and between outlet conduit **16** and exhaust conduit **34**. Upon attainment of the desired vacuum, the RF generator **48** is activated to generate the RF field between electrode plates **46** and therby ionize the gas supplied to the reaction chamber through the intake conduit. the ionized gas or plasma now reacts with the sample. The electrode plates remain energized and gas is continuously passed into and withdrawn from the reaction chamber until the desired degree of reaction between the plasma and the sample is obtained.

To increase the flow of plasma past the sample and thereby obtain an optimal reaction between the two, the inlet and outlet openings **68** and **70** are preferably disposed at about the longitudinal center of container **12** and oppose each other. Gas flow in a longitudinal direction is thereby minimized. Such gas flow is undesirable because an increasing portion of gas, for example oxygen, reacts with the sample. Consequently, as the gas travels along the sample the amount of gas available for reaction with the sample diminishes. As a result, the reaction rate between the plasma and the sample decreases in the direction of travel of the gas. The above placement of inlet and outlet openings **68** and **70**, however, minimizes the longitudinal component of the gas travel past the sample. The opposing positions of the inlet and outlet openings require that all plasma travels transversely through the reaction chamber to assure maximum contact between the plasma and the sample and prevent portions of the plasma from entering the outlet conduit **16** without contacting, or at least coming into the vicinity of the sample.

As compared with prior art plasma generating and reacting machines, the machine of this invention permits the attainment of the desired degree of reaction at substantially shorter times, since the plasma intensity and the gas flow rate can be increased over that possible in the prior art. At the same time, the power requirements of the apparatus are reduced as compared to prior art apparatus of the same capacity since the plasma generating RF field surrounds the sample; and energy losses from a premature plasma breakdown, when the latter is generated remote from the sample, are avoided.

I claim:

1. A reaction chamber assembly for use with a source of gas to be ionized and a radio frequency electrical generator, for reacting ionized gas with a non-gaseous substance, comprising in combination:

an elongate container defining a chamber interiorly thereof and having a gas inlet conduit and outlet conduit, respectively, to receive and expel gas, the ends of said inlet and outlet conduit located to face in the direction of the longitudinal axis of said chamber, and an access opening at one end of said container to receive the substance to be treated in said chamber, said container shaped to have a curved exterior surface and define a relatively constant cross-section along the longitudinal axis thereof;

a removable cover for closing said access opening and sealing said chamber from the exterior thereof;

a pair of opposing electrodes disposed at the exterior of said container and insulated therefrom; wherein a substance received in said chamber is disposed between said electrodes, said electrodes shaped to conformably embrace the curved exterior surface of said container so that the container is slideable between said electrodes in an axial direction;

support means associated with the reaction chamber assembly and secured to said electrodes to support said container in operable position;

a support base to carry said support means;

a supply conduit and an exhaust conduit carried by said support base and positioned so that said supply and exhaust conduit ends engage said inlet and outlet conduit ends, respectively, when the container is slidably placed between said electrodes;

sealing means between the ends of the conduit to seal the interior of the chamber from the exterior of the container; and

means for electrically connecting said electrodes to the radio frequency electrical energy generator, whereby energization of the electrodes by the generator subjects at least a portion of the chamber to an electric radio frequency field to ionize gas in said chamber.

2. A reaction chamber assembly in accordance with claim 1 wherein the electrodes comprise electrode plates spaced from the container, and including an insulating material between and in contact with the container and the plates.

3. A reaction chamber assembly according to claim 1 wherein the sealing means comprise annular, resilient members between ends of the inlet and the outlet conduits, and between the supply and the exhaust conduits, each member being mounted to one of such conduits and positioned to engage the respective mating conduit ends when the container is operatively placed between the plates.

4. A reaction chamber assembly according to claim 1 wherein the container has a longitudinal axis, the electrodes comprise electrode plates extending over the major length of the container, and wherein a surface of the electrodes facing an exterior surface of the container is proximate and substantially parallel to the exterior surface.

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