June 20, 1972 R. L. BERSIN 3,671,195

METHOD AND APPARATUS FOR ASHING ORGANIC SUBSTANCE

Filed Aug. 19, 1968

FIG. 1

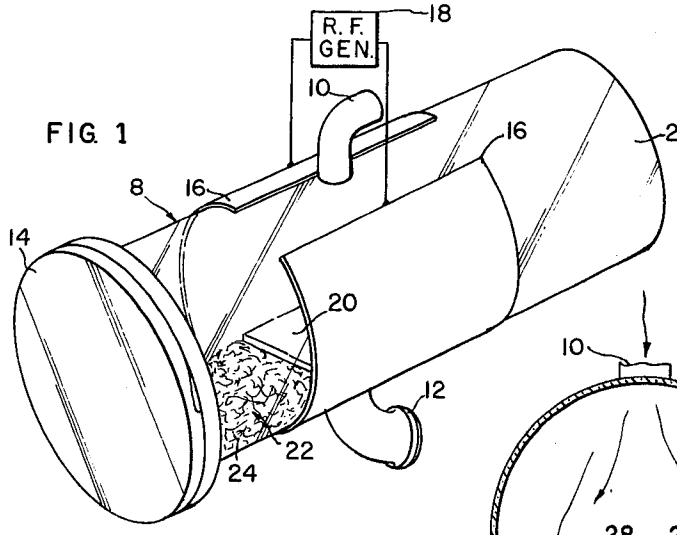


FIG. 6

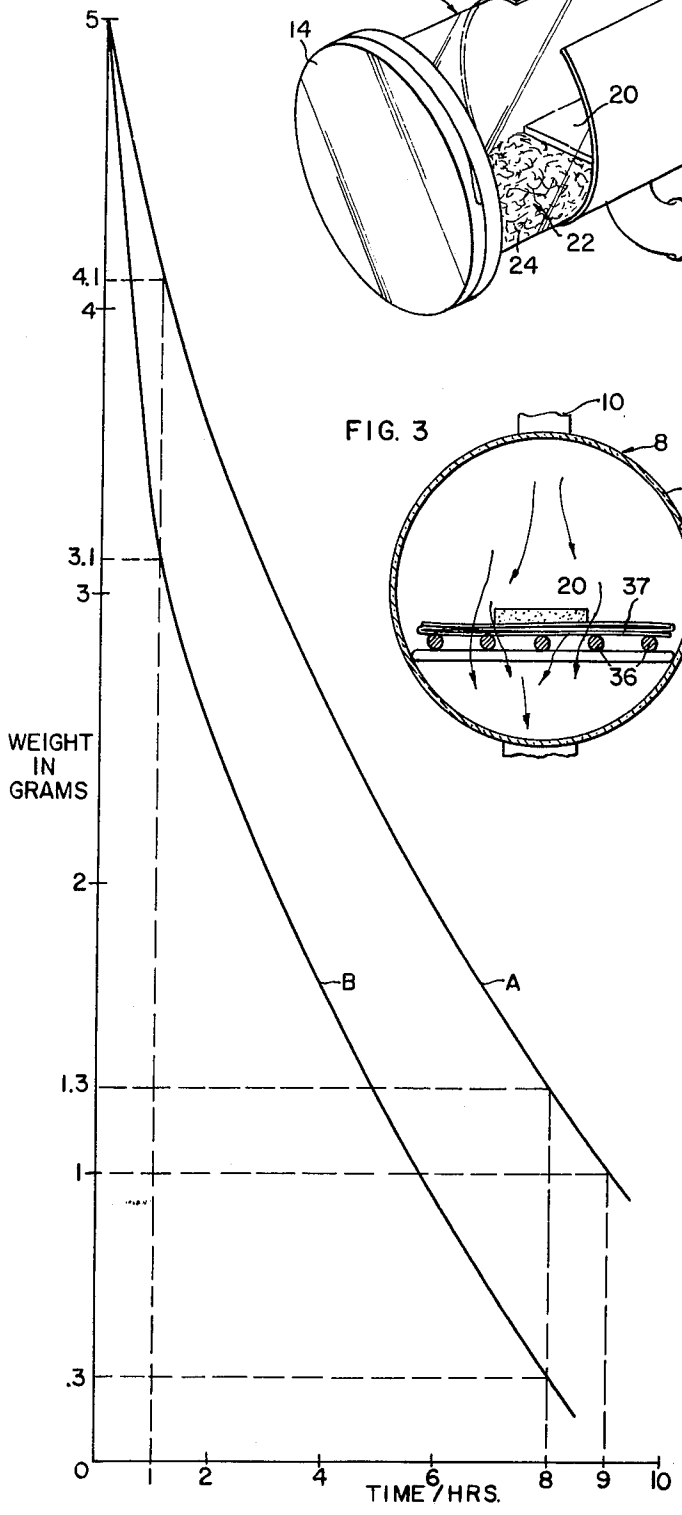


FIG. 2

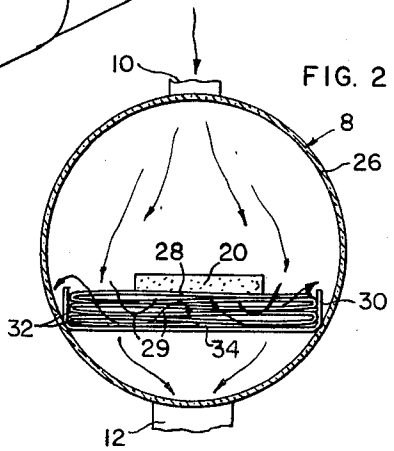


FIG. 3

FIG. 4

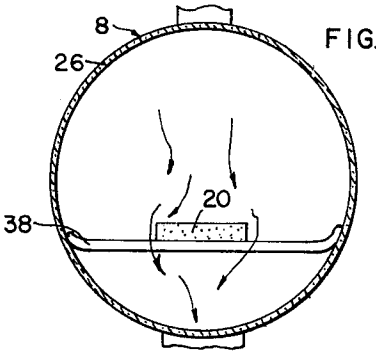
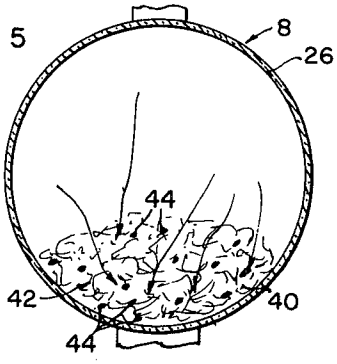


FIG. 5



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METHOD AND APPARATUS FOR ASHING ORGANIC SUBSTANCE

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ABSTRACT OF THE DISCLOSURE

A support structure for a specimen to be treated in the chamber of a plasma machine wherein excited gas flows through the chamber for reaction with the specimen, such structure being fabricated of a fibrous material shaped to be supported inside the reaction chamber and to carry the specimen, the material itself being a substance which is inert to the excited gas yet permeable thereto, whereby the excited gas flows through as well as around the support structure and thus into substantial contact with the surfaces of the specimen to be treated.

BACKGROUND OF THE INVENTION

This invention relates to the low temperature ashing of organic substances and more particularly to an apparatus and a method which permit the simultaneous ashing of all sides of the organic substance.

In various commercial and research processes involving the treatment and/or analysis of organic substances, samples, such as human and animal tissue, plant leaves, coal, etc., are ashed, that is their organic ingredients are oxidized at a low temperature, to obtain an inorganic residue which may then be further processed or analyzed. Exemplary of instances in which organic substances are ashed include pathology laboratories in hospitals in the analysis of human tissue; inspection and quality control centers of drug manufacturers which are legally required to prepare ashed samples of preparations sold by them; and cigarette manufacturers who regularly and as a matter of course ash tobacco and cigarette paper to determine their inorganic contents.

Such ashing may be conducted by plasma generating techniques which are becoming increasingly popular. That is, the sample to be ashed is placed in a stream of an ionized gas of low temperature, commonly referred to as low temperature plasma, for example ionized oxygen (O_2) gas, generated and utilized in equipment generally known in the industry as a plasma ashing machine. The sample to be ashed is placed on a tray or boat positioned in the interior of a reaction chamber of such a machine, and remains there until the ashing is complete.

Prior art trays generally prevent or at least greatly inhibit plasma from contacting that side of the sample which rests on the tray. Consequently, the ashing commences at the surfaces of the sample out of contact with the tray or other supportive structure, and progresses toward the supported and contacting surfaces. This in itself substantially prolongs the required ashing time since the complete oxidation of the sample requires that the ashing progress from the unsupported sides to the supported side through the full thickness of the sample. In other words, at a given ashing rate, the ashing time is that thickness in mm. divided by the ashing rate in mm. per hour. More importantly, already ashed material covers the remainder of the sample which greatly reduces the rate of ashing with an increased build-up of ash on the sample.

SUMMARY OF THE INVENTION

The present invention provides apparatus for the low temperature ashing of organic substances in a stream of an excited gas in which the sample to be ashed is supported by a gas pervious fabric which is inert to the excited gas. The fabric comprises an arrangement of loosely interconnected fibers constructed of an inorganic substance and permits the passage of the gas stream therethrough. The fibers have a sufficient strength to prevent the sample from compacting the loose arrangement of fibers.

In the presently preferred embodiment of the invention, the fibers are quartz fibers and the fabric is supported by a tray or a support structure removably placed in a reaction chamber of the apparatus. The thickness of the fabric is sufficient to permit passage of the excited gas between the sample and the tray so that the ashing of the sample can proceed from all sides of the sample toward the center thereof.

The required time for ashing the sample is thereby greatly reduced since the oxidation proceeds from all sides of the sample. Thus, for a given sample size the required ashing time is theoretically reduced by 50 percent since the ashing time now equals one-half of the thickness of the sample (ashing now proceeds from the supported and the unsupported sides of the sample) divided by the ashing rate in mm. per hour. In addition, the thickness of the layer of ashed material on the sample is reduced, thereby eliminating some of the decrease in the ashing rate from the build-up of ash on the sample. A further advantage may be achieved by constructing the support fabric so that it permits ashed material to gravitationally drop off the bottom side of the sample, thereby enabling virtually free access for the ionized gas to the bottom side which further increases the rate of ashing.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a perspective view of a reaction chamber in which a sample to be ashed is supported by a support fabric constructed according to the present invention;

FIG. 2 is a cross sectional view of the chamber and shows a tray for supporting the fabric;

FIG. 3 is a cross sectional view similar to FIG. 2 but shows an elongate, spaced-apart lattice of rods for supporting the fabric;

FIG. 4 is a view similar to FIG. 3 but shows another preferred arrangement of the fabric;

FIG. 5 is a cross sectional view similar to FIG. 2 and shows fabric supporting a previously powdered sample; and

FIG. 6 is a diagram showing the changes in the ashing time for a sample of a given size as a function of the manner of supporting the sample in the reaction chamber.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

Referring first to FIG. 1, a reaction chamber **8** includes an inlet **10** to which suitable conduits (not shown) are connected to transport gas, such as oxygen gas (O_2), at pressures of a few mm. of mercury from a source (not shown) of gas into the chamber at a closely controlled rate. An outlet **12** is connected to a suction pump (not shown) for exhausting or withdrawing gas from the interior of the chamber. The chamber is closed with a cover **14** and includes means such as opposing plate electrodes **16**, or an inductor coil (not shown), for ionizing the gas inside the chamber. The electrodes are connected with a radio frequency (RF) generator **18** which energizes the electrodes to generate an RF field between them. The gas is excited and ionized by the RF field to form a plasma at a relatively low temperature, for example between about 100° and 150° C., where such plasma generally exhibits a visible glow. When the plasma is transported over and into contact with an organic substance, such as sample **20**, the plasma, under normal conditions of plasma ashing techniques, slowly oxidizes the substances generally with little if any loss of the organic residue, as would be the case with high temperature ashing techniques.

Still referring to FIG. 1, a sample or specimen **20** is disposed in the chamber and rests on a supporting fabric **22** placed on the bottom of the chamber. The fabric comprises a large number of fine fibers **24** which are loosely interengaged, in a manner similar to the interengagement of loose woven fabric fibers, for example, to permit the passage of plasma therethrough while spacing the sample from wall **26** defining chamber **8**. Thus, the plasma present within chamber **8** may reach all surfaces of the sample, including the side of the sample resting on the supporting fabric.

To ash sample **20**, RF generator **18** is activated to energize plate electrodes **16**; and a valve (not shown) is opened to connect the chamber with the source of appropriate gas. A pressure regulator may be interposed between the source and the chamber to maintain constant gas pressure in the chamber, while a suction pump (not shown) withdraws gas from the chamber at a predeterminable rate. The RF field between the electrodes ionizes the gas therebetween, the temperature of the ionized gas rising slightly, and the gas becomes a low temperature, chemically active plasma. Upon contacting the exterior surfaces of the sample, the plasma begins to oxidize, or ash, the sample from such surfaces toward the center. The remaining ash includes all inorganic components of the sample which subsequently may be analyzed.

Referring to FIGS. 1 through 5, it will be observed that the plasma can reach all sides of the sample, including underside **28** resting on supporting fabric **22**. The gas pervious fabric permits the passage of the plasma therethrough and into contact with the underside of the sample as indicated by arrows **29** in FIG. 2.

Although the fabric may be placed directly onto wall **26** of chamber **8**, as illustrated in FIG. 1, it is preferred to provide a support structure such as a tray **30** (shown in FIG. 2) onto which the fabric and the sample can be placed. This facilitates the insertion and removal of the sample and the fabric into and out of the chamber. Quantative analysis can then be performed by weighing the whole tray, including the ash and the fabric, without danger of losing parts of the ash in the reaction chamber.

Referring to FIGS. 2 and 3, in a presently preferred embodiment of this invention the support fabric is constructed in the form of mats or sheets **32** which are placed into tray **30** and stacked to space the sample from floor **34** of the tray. Alternatively, fabric mats **32** can be placed on a lattice or grid defined by elongate, spaced apart rods **36** disposed in the reaction chamber as illustrated in FIG. 3. The sample is thereby spaced from chamber walls **26**. The open spaces of the grid or rod lattice permit passage of the plasma as it travels from inlet **10** to outlet **12** and facilitates the gas exchange in the space beneath underside **28** of sample **20**. This enhances the obtainable rate of oxidation of the sample.

Referring to FIG. 4, mats **38** may be supported directly by wall **26** of the reaction chamber by bonding the mat to the walls or mechanically securing the mat thereto. Mat **38** also spaces the sample from the chamber wall and permits the free passage of ionized gas past and around the sample as well as through the mat in a manner substantially similar to the grid or lattice arrangement shown in FIG. 3.

Referring to FIG. 5, if the sample has been pulverized prior to its ashing, as is commonly the case in pathology laboratories, for example, the complete ashing of the sample is often difficult to achieve and the ashing rate is low since the plasma cannot readily penetrate into the center of the pulverized mass past the ashed layer on the sample. To shorten ashing time in such cases, the present invention provides that fibers **40** be loosely bunched together to form a fiber wool pile **42** having many open spaces through which plasma can penetrate. The pulverized sample **44** can be dropped into such spaces to be supported by one or more fibers within the interior of the fiber wool pile. The sample divides into small particles which are positioned throughout the fiber wool as shown in FIG. 5. Plasma enters the fiber wall and directly contacts each particle of the sample without having to go through or past ashed material before reaching non-oxidized portions of the sample. Ashing times are thereby significantly reduced.

It is, of course, necessary to construct the fibers supporting the samples of a material which is inert to the plasma; that is material which does not oxidize or is otherwise attacked by the ionized gas in the reaction chamber. For ashing, a non-oxidizable inorganic material capable of withstanding the temperatures to which it is exposed in the reaction chamber is required. In addition, the material must have sufficient strength or stiffness to support the sample without crumbling or compacting under the load of the sample which would reduce the accessibility of plasma to portions of the sample.

Silicon containing materials such as glass, for example, are particularly well suited for supporting organic samples during ashing because they combine the required inertness to the chemically active plasma with wide availability and relatively low costs. To obtain a small diameter, fine fibers, which reduce the contact area between the sample and each fiber and thereby increases the area of the sample exposed to the plasma without obstruction, best results have been achieved with fibers constructed of quartz. Quartz fibers, moreover, have an excellent ability to hold their shape, particularly if formed into mats as illustrated in FIGS. 2 through 4, to maintain a stream of plasma past the supported side of the sample. When desirable or necessary, other materials can, of course, be substituted for the above described.

Referring to FIG. 6, the efficacy of the invention and the reduction in the ashing time for a sample of dried kidney weighing 5 grams is diagrammatically illustrated. Curve A shows the progress of the ashing when the sample is placed on a conventional tray in accordance with the prior art. After approximately nine hours, there remained one gram of unashed material. Curve B represents the ashing progress of an identical sample placed on a support fabric constructed of quartz fibers and formed into mats, as illustrated in FIGS. 2 through 4, after eight hours of ashing, there remains a non-oxidized residue of merely 0.3 gram, a substantial improvement over the prior art. The greater efficiency of the ashing process when utilizing support fabrics according to the present invention is perhaps more drastic when considering that after one hour of ashing according to the prior art (curve A) only 0.9 gram of the sample has been ashed, while 1.9 grams of the sample have been ashed when supported by the fibrous

fabric. Thus the ashing rate of the sample has more than doubled.

The foregoing description of my invention may also be summarized as follows insofar as a new and useful method is disclosed for the practice of plasma ashing of various types of specimens in a stream of excited gas passing through a reaction chamber.

(1) Selecting a material inert to the excited gas;
(2) Placing a gas previous arrangement of the material in the reaction chamber so that the arrangement defines a supporting surface for receiving a specimen to be ashed and positions the specimen in spaced relation to the wall of the chamber;
(3) Placing the specimen onto the supporting surface; and
(4) Passing the excited gas into contact with the specimen through the gas pervious arrangement of the gas-inert material so that the specimen is evenly and simultaneously ashed over substantially its full exterior surface.

In addition to the foregoing description of the process embodying my invention, a further advantage of even greater increased ashing rate may be achieved by constructing the support fabric so that it permits ashed material to gravitationally drop off the bottom side of the specimen being ashed. In this way the portion of the ashed specimen near the surface of the support fabric or material is removed from the main body of the specimen thus enabling virtually free access for the ionized gas stream to theretofore unashed portions of the bottom side of the specimen, which further increases the rate of ashing.

It will be understood that the advantages of the present invention are independent of the plasma generating and reacting apparatus employed. Thus, plasma can be generated between electrode plates in the space surrounding the sample, as shown in FIG. 1, or it may be generated in an ionization chamber (not shown) communicating with and spaced from the reaction chamber.

I claim:

1. A support member for a specimen to be treated in the chamber of a plasma machine wherein excited gas flows through the chamber for reaction with the specimen, said member comprising:
a tray supported by the walls of the chamber for positioning and supporting material disposed therein in spaced relation to the walls; and
a fibrous material fabricated of a substance inert to the excited gas and permeable thereto, said fibrous material comprising fibers interengaged to form an irregularly shaped body of bunched fibers supported by said tray for carrying the specimen to be treated whereby the excited gas may be caused to flow past and through the fibrous material and into substantial contact with the surfaces of the specimen supported thereon and to be treated.

2. The combination according to claim 1 wherein a portion of the tray supporting the fibers includes openings permitting passage of the gas stream therethrough.

3. A support member for a specimen to be treated in the chamber of a plasma machine wherein excited gas flows through the chamber for reaction with the specimen, said member comprising:
a substantially flat support structure defined by spaced apart, elongate members supported by the walls of the chamber and permitting passage of the gas stream through the open spaces intermediate the members; and
a fibrous material fabricated of a substance inert to the excited gas and permeable thereto, said material

shaped to be supported by said support structure and to carry the specimen, said support structure positioning and supporting the fibrous material in spaced relation to the walls, whereby the excited gas may be caused to flow past and through the fibrous material and into substantial contact with the surfaces of the specimen to be treated.

4. A method for ashing specimens in a stream of excited gas passing through a reaction chamber comprising the steps of:
(a) selecting a material inert to the excited gas,
(b) placing a gas pervious arrangement of the material in the chamber, the arrangement defining a supporting surface for receiving a specimen and positioning the specimen in spaced relation to the walls of the chamber,
(c) placing the specimen onto the supporting surface, and
(d) passing the excited gas into contact with the specimen and through the gas pervious arrangement of the material, whereby the specimen is evenly and simultaneously ashed over substantially its full exterior surface.

5. A method according to claim 4, including the step of gravitationally removing at least a portion of the ashed specimen from the body of the specimen by positioning the supporting surface so that such portion of the ashed specimen proximate the supporting surface is gravitationally separated from the specimen.

6. A support member for a specimen to be treated in the chamber of a plasma machine wherein excited gas flows through the chamber for reaction with the specimen, said member comprising:
a tray supported by the walls of the chamber for positioning and supporting material disposed therein in spaced relation to the walls; and
a fibrous material fabricated of a substance inert to the excited gas and permeable thereto, said fibrous material comprising flat fiber mats arranged in a plurality of layers supported by said tray for carrying the specimen to be treated, whereby the excited gas may be caused to flow past and through the fibrous material and into substantial contact with the surfaces of the specimen supported thereon and to be treated.

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E. A. KATZ, Assistant Examiner

U.S. Cl. X.R.

23—253 PC, 230 B; 204—312; 313—231; 315—111; 317—4