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Kaartinen

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[54] **COMBUSTION APPARATUS AND METHOD FOR MATERIALS CONTAINING A RADIOACTIVE ISOTOPE TRACER**

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[51] **Int. Cl.** **G01n 31/00**

[58] **Field of Search** .... 23/253 PC, 230 PC; 250/106 T, 83 SA

[56] **References Cited**

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3,058,814	10/1962	Bennet	23/253 PC
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3,485,565	12/1969	Kaartinen	431/3

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[57] **ABSTRACT**

An apparatus and method are provided for the combustion of starting materials containing radioactive isotope tracers. Starting material contained in a special cup-shaped holder is placed in an ignition basket and is burned in a combustion chamber. The combustion products are continuously exhausted from the combustion chamber and passed into an isotope recovery system. An ignition promoter may also be provided to assist in ignition of the cup-shaped holder and the starting material contained therein.

9 Claims, 3 Drawing Figures

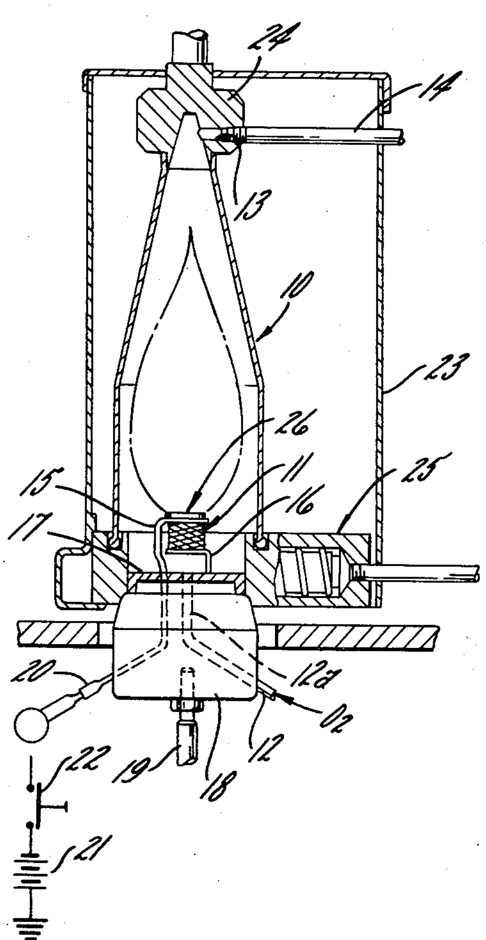

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FIG. 1.

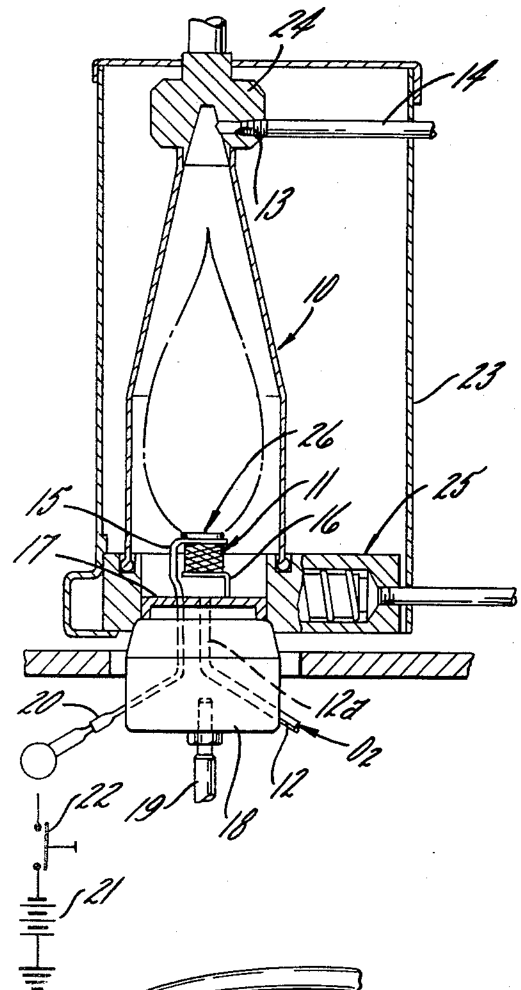
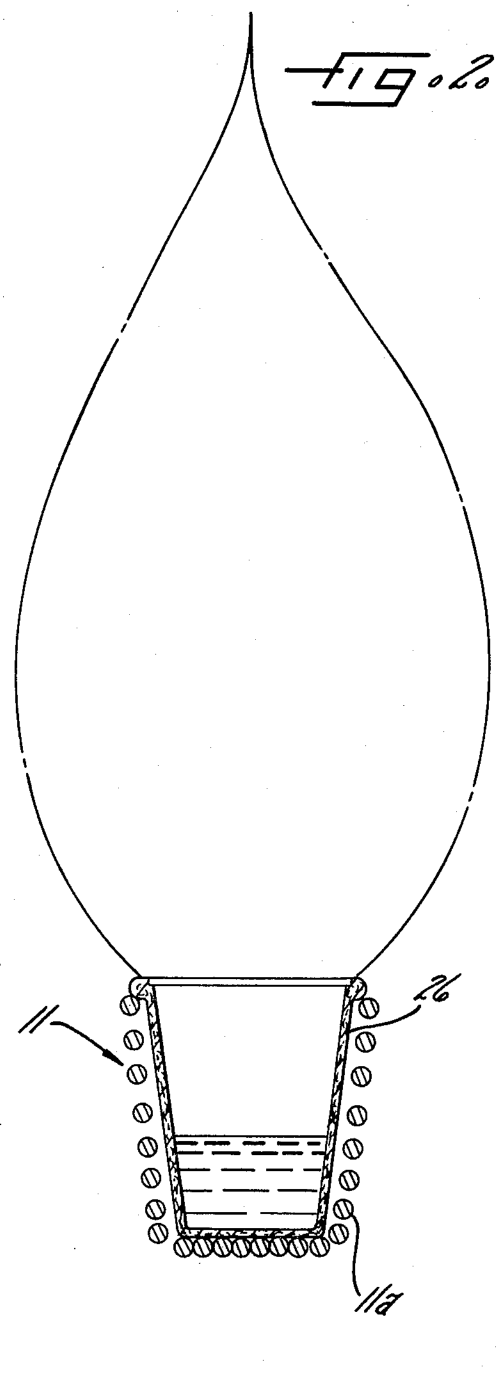


FIG. 2.



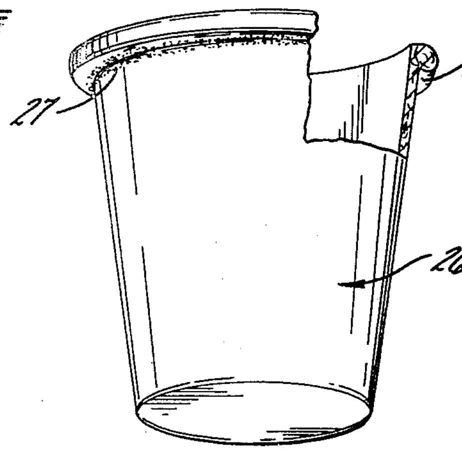


FIG. 3.

COMBUSTION APPARATUS AND METHOD FOR MATERIALS CONTAINING A RADIOACTIVE ISOTOPE TRACER

DESCRIPTION OF THE INVENTION

The present invention relates generally to the combustion of starting materials containing a radioactive isotope tracer in the preparation of fluid samples for radioactive isotope tracer studies. More particularly, the invention relates to an apparatus and method for combustion of liquid or solid starting materials from which fluid samples may be prepared for radioactive isotope tracer studies.

Various combustion techniques have been devised for the preparation of samples for radioactive isotope tracer studies. One particularly efficient and commercially attractive technique for such sample preparation involves combustion of a starting material containing one or more isotope tracers, recovery and liquefaction of the gaseous combustion products containing the isotopes, and isolation of the individual isotopes to permit their quantitative determination.

Combustion techniques of this general type (combustion-recovery-isolation) have been disclosed in my U.S. Pat. Nos. 3,485,565 and 3,542,121, and in my copending applications Ser. No. 728,939, filed May 14, 1968, now abandoned in favor of my continuation application Ser. No. 277,261, filed Aug. 2, 1972, Ser. No. 58,635, filed July 27, 1970; Ser. No. 820,269, filed Apr. 29, 1969, now U.S. Pat. No. 3,682,598; and, Ser No. 242,481 now U.S. Pat. No. 3,761,228 filed Apr. 10, 1972; as well as in their foreign counterparts. These techniques have involved manual placement by an operator or technician of an isotope tracer containing material into the apparatus for combustion, followed by various manipulations necessary for starting, controlling and terminating flows of the gases and liquids used in preparing the samples.

In the prior combustion techniques, it has usually been desirable that the starting material to be burned be in solid form. Typically, the starting material has been wrapped in an analytically pure carrier material, such as special low ash filter paper, and then pelleted. The resulting pellet has then been placed within an ignition basket. Combustion of the pelleted starting material has been achieved by passing electric current through the ignition basket, and admitting gaseous oxygen into the combustion chamber surrounding the basket so that the pyrolysis of the pelleted starting material takes place.

Desirably, in order to achieve complete combustion of the starting material, all of the vapors and combustion products from the starting material should pass through the combustion flame, since only the main flame above the ignition basket is hot enough to insure complete combustion. Also, it is desirable that the rate of combustion not exceed the rate of supply of gaseous oxygen.

It has been found that prior combustion techniques sometimes fail to achieve the desirable combustion goals in various respects. Often the heat of ignition or heat of combustion of the pelleted starting material is inadequate to fully pyrolyze the material during the time that the pellet is surrounded by the combustion flame. On the other hand, if the heat of ignition or heat of combustion is adjusted to provide very fast pyrolysis,

the tendency is for flame jets of the pyrolyzation products to shoot sidewise and downwardly (rather than upwardly) where there is not enough space or heat to achieve complete combustion. When this occurs, a positive feedback is created between the rate of combustion and the rate of pyrolyzation, thereby continuously increasing the demand for oxygen until there is insufficient oxygen for complete combustion. Only when the main part of the flame remains above the starting material is this positive feedback prevented so that the rate of combustion may be controlled, for example by controlling the heat input to the ignition basket. When pyrolyzation products escape complete combustion the isotope recovery is incomplete, resulting in undesirable "memory" in the apparatus and consequent spillover errors adversely affecting the analytical results.

In attempting to minimize these problems when using the filter paper pellet technique, it is necessary to use a large quantity of wrapping paper in relation to the quantity of starting material, with the result that the pellet technique can accommodate only very small quantities of starting materials. Starting materials which are in liquid form present different problems. It has been found that fast burning liquid or plastic starting materials cannot be successfully burned using the pellet technique. Slow burning liquid starting materials, for example which include water as a main constituent, require an inordinately large quantity of carrier material. Typically, the weight of filter paper used in such cases must be greater than the weight of the starting material. Again, the usable starting material size is considerably reduced. With all of these prior techniques, the necessary step of pelletizing is bothersome and time consuming.

It is a primary object of the invention to provide an improved combustion method and apparatus for materials containing a radioactive isotope tracer which eliminates the need for pelleting. An allied object is to provide such a method and apparatus which work as efficiently with liquid as with solid starting materials, and which in either case affords complete combustion of these materials within the combustion flame.

Yet another object of the invention is to provide an improved combustion apparatus which may incorporate a non-noble metal type, and therefore structurally stronger, ignition basket than has been useful in the past without sacrificing any speed or efficiency of combustion.

It is a still further object of the present invention to provide an improved method and apparatus which significantly further reduce the combustion time below that required using my aforementioned previously disclosed techniques, with a corresponding meaningful increase in the isotope containing sample preparation rate.

Other objects and advantages of the invention will become apparent from the following detailed description and upon reference to the accompanying drawings, in which:

FIG. 1 is a schematic diagram of a combustion apparatus embodying the present invention, for use in the preparation of samples for radioactive isotope tracer studies (flame shown in dotted lines);

FIG. 2 is an enlarged, elevational cross-sectional view of an illustrative ignition basket and cup-like holder for the starting material to be burned in the combustion apparatus of the invention (flame shown in dotted lines); and,

FIG. 3 is a further enlarged perspective view, partially cut away, of the illustrative cup-like holder for the starting material shown in FIGS. 1 and 2.

While the invention will be described in connection with certain preferred embodiments, it will be understood that it is not intended to limit the invention to these particular embodiments. On the contrary, it is intended to cover all alternatives, modifications and equivalent arrangements as may be included within the spirit and scope of the invention as defined by the appended claims.

Turning now to the drawings, in FIG. 1 there is illustrated a combustion apparatus 10 for use in the preparation of samples for radioactive isotope tracer studies, such as studies involving tissue distribution and residue levels of drugs in plants and animals. In the preparation of such samples, the starting material containing the radioactive isotope tracer, such as plant or animal tissue, is burned to convert the carbon in the starting material to carbon dioxide and the hydrogen to water, and the radioactive isotope tracer is then recovered from the resulting combustion products. The details of the sample recovery are described in my aforementioned patents and applications and will not be repeated here. Suffice it to say that the combustion chamber 10 is followed by various units, interconnecting conduits, and associated apparatus for treating the gaseous combustion products to separate and effect recovery of each radioactive isotope tracer. The apparatus in this instance is particularly adapted for recovery of ^3H and ^{14}C isotope tracers.

With reference to FIG. 1, material containing the radioactive isotope tracers, in either solid or liquid form, is placed in an ignition basket 11 within the combustion chamber 10. The apparatus operator thereupon pushes a start button for a pre-programmed pneumatic control unit (not shown), and combustion oxygen is admitted to the chamber 10 through an oxygen inlet 12. The isotope containing material is quickly ignited, and the resulting gaseous combustion products pass upwardly through the combustion chamber 10 and then through an exhaust port 13. The combustion gases continue through a conduit 14 and into the recovery system (not shown). Following recovery, a tritium sample collection vial and a carbon sample collection vial (neither shown) may be removed from the recovery system by the apparatus operator, and the radioactivity of the tritium and carbon samples determined using a photomultiplier or other suitable radioactivity counting device.

While various combustion chamber designs may be used in practicing the invention, it is preferred to use a generally flame shaped combustion chamber or flask of the type disclosed in my U.S. Pat. No. 3,485,565, or in my copending U.S. applications Ser. No. 820,269, filed Apr. 29, 1969, now U.S. Pat. No. 3,682,598 and Ser. No. 242,481, filed Apr. 10, 1972.

The combustion chamber 10 illustrated in FIG. 1 is of this general type. The radioactive isotope containing starting material to be burned is placed in the ignition basket 11 which forms a part of the electrical ignition system. To this end the basket 11 is made of nichrome or like electrically resistive material, so that it functions as an electrical resistor in the ignition system. A pair of electrical conductors 15 and 16 extend upwardly from a mounting plate 17 to support the basket 11 at its upper and lower ends, while also making electrical contact with the basket to connect it into the electrical ignition system. The conductors 15 and 16 extend vertically down through the plate 17 and terminate in depending connector pins beneath it. In order to facilitate the loading of successive specimens for combustion, the mounting plate 17 is supported on a platform 18 which is threaded or otherwise secured onto the end of a pneumatic piston rod 19, which rod forms a part of a retracting and elevating mechanism (not shown) which automatically moves the ignition basket 11 and mounting plate 17 from an open position easily accessible to the operator (not shown in the drawings) to a closed or sealed position within the combustion chamber (as shown in FIG. 1).

For the purpose of igniting a starting material contained in the basket 11, the connector pins depending from the plate 17 are received by a complementary electrical receptacle 20 in the platform 18. The receptacle 20, in turn, is connected to an electrical igniter circuit including a power source such as battery 21 and an ignition switch 22 for applying an electrical voltage across the basket 11, which serves as a resistive type heating element in the igniter system. Thus, the radioactive starting material is ignited by simply closing the switch 22, which is opened again as soon as combustion has been completed.

For combustion of the specimen contained in the basket 11, pure oxygen is supplied to the combustion chamber 10 through the conduit 12 and a cooperating passageway 21*a* formed in the platform 18 and the plate 17. The gas discharge passageway 12*a* in the plate 17 is positioned directly beneath the center of the basket 11, so that the oxygen is fed directly into the combustion zone. The oxygen flow rate is initially adjusted, by suitable valving and flow metering means (not shown), to a level slightly above that required to support combustion of the sample, so that there is a slight excess of oxygen within the combustion chamber 10. This excess oxygen rises through the combustion chamber 10 and is exhausted from the combustion chamber along with the combustion products through the lateral exit 13 at the top of the chamber. A pressurized oxygen supply container (not shown), which functions much like a capacitor, is preferably employed, so that as the demand for oxygen in the combustion chamber 10 diminishes during combustion of the specimen, the oxygen feed to the combustion chamber diminishes accordingly.

The combustion chamber 10 is preferably open at its upper end, with its sidewalls extending upwardly and inwardly above the specimen basket 11 so as to approximate the shape of the flame of a burning sample. This arrangement advantageously minimizes the volume of oxygen-rich atmosphere around the flame, and the walls of the combustion chamber 10 are preheated so as to maintain the wall temperature above the condensation temperature of the vapors contained in the combustion products. With this design, the combustion products tend to be swept directly into the exit 13, with the rising layer of oxygen-rich atmosphere along the chamber sidewalls tending to isolate the combustion products from the sidewalls. Moreover, any combustion products that do contact the chamber walls remain in the gaseous state, even during initiation of the combustion, because the walls are pre-heated and maintained at a temperature above the condensation temperature.

Thus, in the illustrative embodiment of the combustion chamber 10 illustrated in FIG. 1, the walls of the combustion chamber extend vertically upwardly past the ignition basket 11, and then slope inwardly above the basket so as to approximate the shape of the flame represented in broken lines. Surrounding the combustion chamber 10 is a rectangular enclosure 23 which defines an air filled cavity around the outer surface of the chamber 10. For proper location of the combustion chamber 10 within the enclosure 23, the upper end of the chamber meshes with a complementary mounting element 24.

Prior to ignition of the specimen contained in the basket 11, the air in the cavity between the combustion chamber 10 and the enclosure 23 may be heated by means of a heating coil (not shown) located inside a steam generator 25. The air distributes this heat along the walls of the combustion chamber 10 so that the walls are uniformly heated to a temperature above the condensation temperature of the vapors contained in the combustion products to be produced. It has been found that the pre-heating of the combustion chamber walls to maintain the combustion products in gaseous form even during ignition, combined with the flame-shaped configuration of the chamber, permits the combustion products to be exhausted from the combustion chamber, on a continuous basis, so efficiently that there is virtually no residue of combustion products deposited on the chamber walls. The illustrative system also prevents condensation within the exit 13 of the combustion chamber 10, since the exit is also surrounded by the pre-heated air in the cavity between the combustion chamber and the surrounding enclosure 23.

In carrying out the invention an improved combustion apparatus is provided in which the starting material is burned in an open cup-like holder 26 which is placed inside the ignition basket 11, with the holder 26 being formed of a material which permits it to slowly and evenly burn along with the starting material. In this way, means is provided for the products of combustion of the starting material to pass upwardly through the open mouth of the cup-like holder 26 and directly up and through the flame extending above the ignition basket 11. The holder is shown in FIG. 2 containing a liquid starting material to be burned.

As shown, the holder 26 is generally thimble-shaped, although other shapes providing an open top may also be employed such as a cylindrical contour. The holder 26 is preferably made from low ash paper, and is formed from a water slurry of paper pulp using conventional techniques. The thimble-shaped contour for the holder 26 has the advantage that the tapered side walls permit easy removal of the thimble from a forming mold. Also the tapered walls tend to force the starting material contained in the holder to desirably burn from the top downwardly, thus encouraging complete combustion. Suitable sizing materials may be added to render the paper holder waterproof or at least water resistant so that liquid starting materials may be accommodated. Generally, the holder 26 is sized and shaped to fit snugly within the ignition basket 11.

It may be desirable to apply an ignition promoting substance, which may have an ignition point of about 95° C., such as finely powdered phosphorus sesquisulphide (P_4S_3), around the underside of a rim 26*a* which may be provided around the mouth of the holder 26. As shown in FIG. 3, the powdered ignition promoting substance 27 is applied to the underside of the rim 26*a*. The ignition promoting substance functions to assure that the flame will extend entirely around and above the mouth of the holder 26 by the time that the temperature inside the holder rises even a few degrees C. above room temperature. In this way the rim of the holder 26 is within the flame before the temperature of the starting material is significantly increased, so that the pyrolysis products coming off of the starting material pass through the flame.

When the holder 26 containing a starting material is ignited, the upwardly opening mouth of the holder offers an open pathway for the pyrolyzation products to rise into the flame above the ignition basket 11, where the combustion is most complete. In this way the strong positive feedback conventionally encountered between pyrolyzation and combustion is broken, and the rate of combustion may thus be controlled by controlling the temperature of the combustion basket 11. Also, no time is wasted because the starting material begins to burn immediately since there is no carrier material surrounding the starting material. Yet the starting material begins to pyrolyze only after the flame from the ignition promoter and from the combustion of the upper parts of the holder 26 has been established producing radiant heat. Thereby all of the pyrolyzation products from the starting material pass directly through the flame. The holder 26 itself tends to burn at a rather slow, steady rate. Later in the combustion process, the flame surrounds the entire holder 26 to burn the residue of starting material completely.

Where an ignition promoting substance 27 is employed with the holder 26, the holder containing a starting material will ignite in the combustion chamber 10 within a few seconds time and before the temperature within the holder has risen more than a few degrees C. Once the ignition starts, it propagates in one second or less around the mouth of the holder 26 to join above the holder as a dome-like flame. The amount of ignition promoting substance employed is preferably so small that if it were ignited in air (i.e., outside the combustion chamber) the produced heat would not be great enough to propagate the flame to the holder. In other words, the ignition promoting substance functions to promote ignition of the starting material only in a gaseous oxygen atmosphere, in which atmosphere ignition takes place automatically and immediately before any combustible vapors can prematurely escape from the holder to possibly form a potentially explosive or otherwise dangerous mixture.

If desired, other ignition promoting techniques may be used in lieu of the low kindling temperature substances 27 described above. For example, a high voltage electric spark or an infra-red beam directed against the mouth (or rim 26*a* around the mouth) of the holder 26 may be used to promote ignition of the holder and its contents.

Through use of the present invention the combustion of water containing starting materials is greatly improved. Inasmuch as water will not itself sustain combustion it usually will be desirable to add a fuel-like substance such as an alcohol, for example butanediol, to such a starting material. Since both the liquid starting material and the added alcohol vaporize at the same uniform rate and in the same manner, the proportions of each may be determined by their respective vapor pressures. The proportion of the additional fuel can be easily found by starting from low concentrations and increasing the concentration continuously until the flame becomes self-sustaining under the circumstances of a given combustion. If the added fuel is consumed too fast compared to water, the boiling point is too low. If the additional fuel is consumed too slowly, a liquid with a higher vapor pressure should be selected. Alcohols and other liquids containing oxygen are preferable because they burn without sooting.

Solid fuels used in conventional wrapping and pelletizing techniques rarely have a low enough boiling point to vaporize simultaneously together with water to produce even a minimally continuous combustible mixture. They can be pyrolyzed only with their own heat of combustion, which requires more energy than for vaporization of a liquid. With fast and very fast burning starting materials this situation is reversed using the instant invention. Now it is possible to add water to moderate downwardly somewhat the combustion speed of the very fast burning materials, mainly liquids. This is seldom necessary, however, because the vicious circle encountered in conventional techniques resulting in rapidly accelerating, uncontrolled combustion has been broken through use of the open mouth holder **26** of the present invention.

In one typical example, the sample holder **26** may have the following dimensions and characteristics. The holder may be formed from a paper pulp slurry which is pure from a radioactivity measurement standpoint. The holder **26** may have a height of about 20 millimeters, an average width of about 14 millimeters, a wall thickness of about 2 millimeters or less, and may have about 1.5 to 2 milliliters internal volume. It may weigh about 200 milligrams.

One of the advantageous features of the invention is that it is no longer necessary to employ expensive platinum or platinum alloys as materials of construction for the ignition basket **11**. The use of these metals has been necessary in the past in order to catalyze ignition and combustion of the starting material. By utilizing the present invention it is possible to employ much less expensive metals as materials of construction for the ignition basket. That is, efficient and complete combustion of the starting material may be achieved in the apparatus of the present invention by forming the ignition basket of relatively inexpensive high temperature resistance wire. Examples of such wire include "Kanthal," a high electrical resistance chromium-aluminum-cobalt alloy, and "Nichrome," a nickel-chromium-iron alloy.

Not only is the high resistance wire advantageously cheaper to use for the ignition basket **11** than platinum or a platinum alloy, it is much more stress tolerant. Thus it is possible to achieve an improved ignition basket construction by closely spacing the wire coils **11*a*** at and near the base of the basket **11** to restrict oxygen access to the starting material holder **26** at those locations, and by widely spacing the coils **11*a*** in the upper portion of the basket. In this way, the wire coils **11*a*** are spaced so that the access of oxygen to the burning starting material is progressively reduced from the top to the bottom of the basket (see FIG. **2**). This arrangement, in which the wire coils **11*a*** are wound tightly together at the bottom of the ignition basket and loosely at the top, is not feasible with platinum and platinum alloys because of their lack of structural strength. It further facilitates the combustion taking place from the top downwardly through the starting material, i.e., the tightly wound bottom of the basket desirably forces the flame upwardly into the space above the ignition basket **11** and the holder **26** with the result that combustion takes place from the top downwardly through the starting material placed in the holder.

Platinum has a further disadvantage in that it has a high thermal resistivity coefficient. This tends to cause a platinum ignition basket to jump to full temperature very quickly, resulting in a high beginning flame and unduly speeding up the initial part of the combustion cycle, when the heat of the basket is mostly needed for completion of combustion at the end of the cycle. Through use of the present invention, the basket heat is present when it is most needed during the combustion cycle.

One of the advantages of the invention is the saving that results from elimination of the carrier material heretofore required, and the consequent increase in permissible size of the starting material. Inasmuch as the pyrolyzation products freely escape through the mouth of the holder **26** of the present invention, the walls of the holder may be made very thin and yet will prevent the pyrolyzation products from escaping in other directions than upwardly through the flame.

I claim as my invention:

1. An apparatus for the flame combustion of a starting material, comprising: a combustion chamber, an ignition basket in said combustion chamber, means for supplying gaseous oxygen to said combustion chamber, means for holding the starting material to be burned, said holding means adapted to be placed within said ignition basket and having a mouth opening upwardly into the space above said ignition basket, and said holding means including an ignition promoting substance arranged around said mouth.

2. An apparatus as defined in claim **1** in which said holding means is a thimble-shaped cup opening upwardly into the space above said ignition basket.

3. An apparatus as defined in claim **1** in which said ignition basket is formed of non-noble electrically resistive material.

4. An apparatus as defined in claim **3** in which said non-noble electrically resistive material comprises wire having relatively high electrical resistivity, said wire having coils spaced relatively widely apart at the upper end of said basket and relatively closer together toward the lower end of said basket for insuring that access of oxygen to the starting material in said holding means is progressively reduced from the top to the bottom of said basket.

5. The method of burning a starting material in a combustion chamber so as to permit collection of the products of combustion for subsequent processing, said method comprising the steps of:

a. forming a thimble-shaped cup-like holder of combustible material having a mouth at its upper end;

b. positioning an ignition promoting substance around the mouth of the holder;

c. positioning the starting material to be combusted in the holder;

d. positioning the holder in an open ignition basket within the combustion chamber;

e. supplying gaseous oxygen to the combustion chamber; and,

f. supplying heat to the ignition basket and the holder for completely burning the holder and the starting material contained therein.

6. A method as defined in claim **5** wherein access of oxygen to the burning starting material within the combustible holder is progressively reduced from the top to the bottom of the ignition basket.

7. A method as defined in claim **5** in which the starting material contains at least one radioactive isotope.

8. A method as defined in claim **5** in which the starting material is a liquid.

9. A method as defined in claim **5** in which the starting material is a solid.

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