May 1, 1956 P. H. SHERRICK 2,743,995

METHOD OF SAMPLE BURNING FOR MICRO-CHEMICAL COMBUSTION ANALYSIS

Filed Oct. 2, 1952 3 Sheets-Sheet 1

Fig. 1.

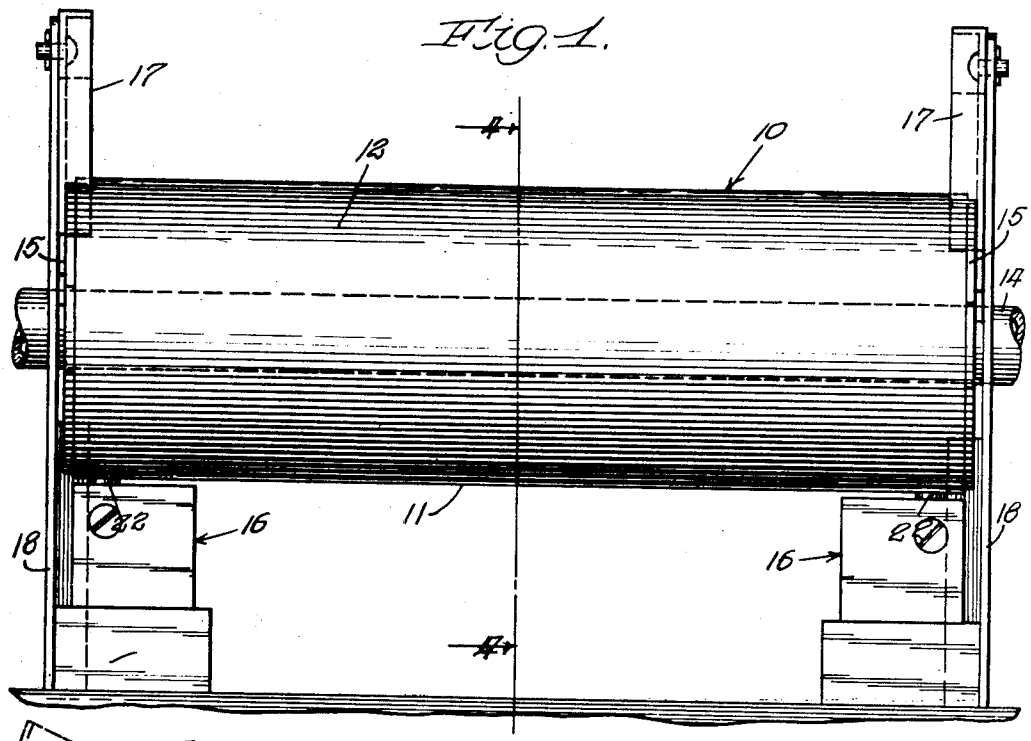

Fig. 2.

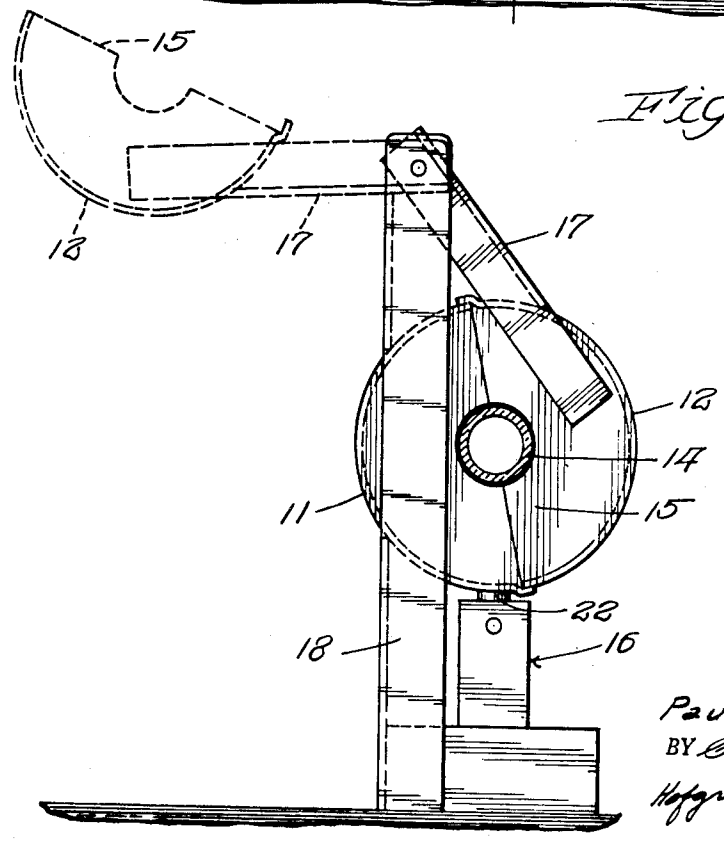

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Fig. 3.

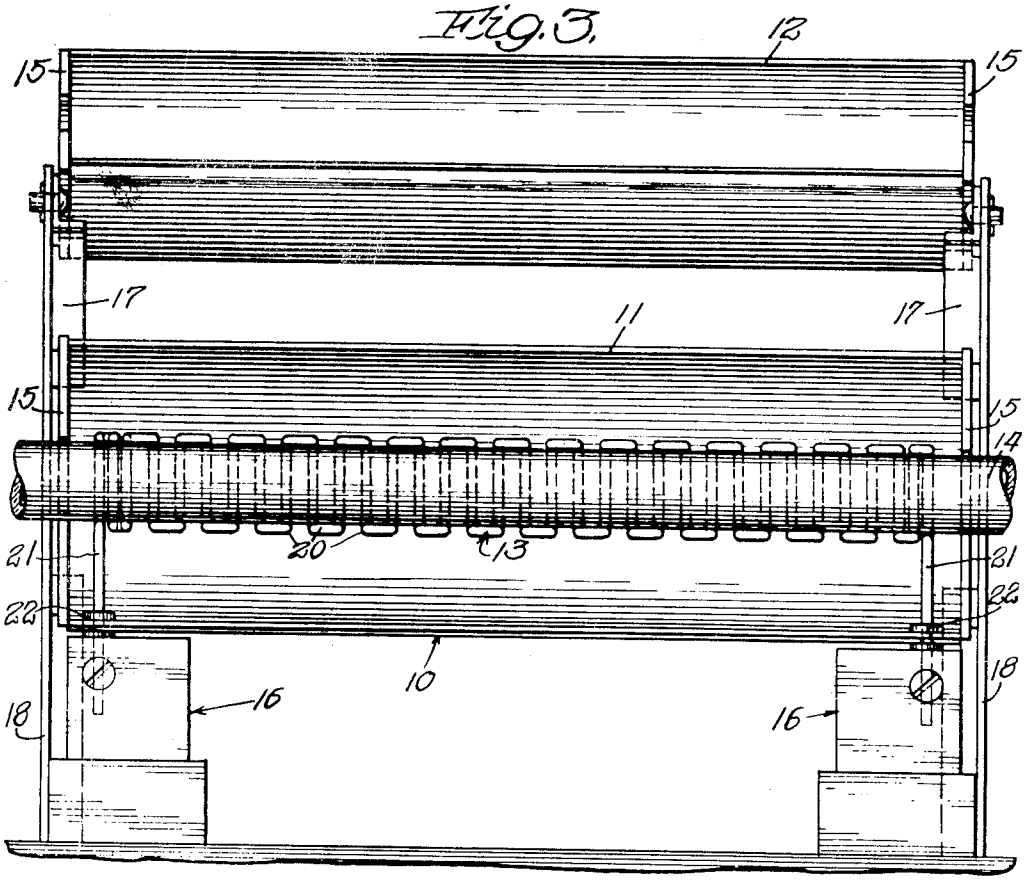


Fig. 4.

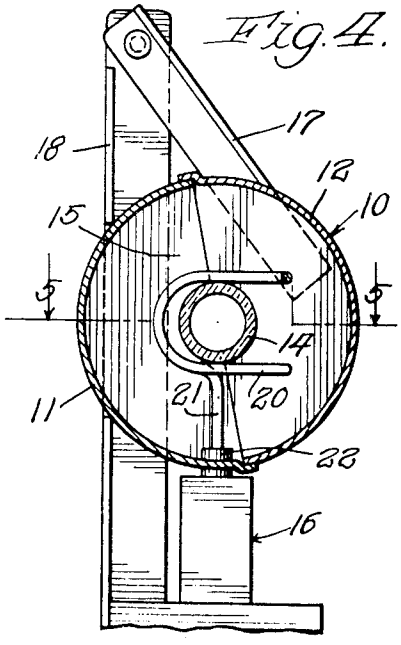
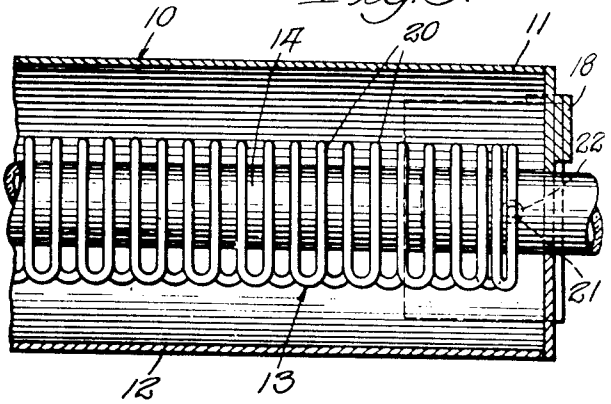


Fig. 5.



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Filed Oct. 2, 1952 3 Sheets-Sheet 3

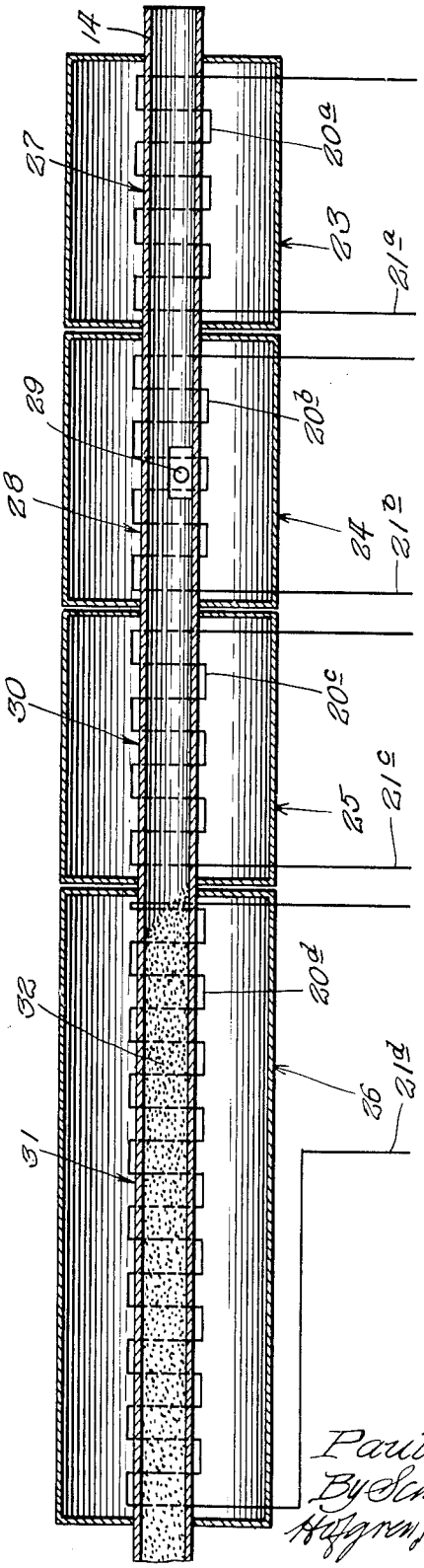

Fig. 6.

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METHOD OF SAMPLE BURNING FOR MICRO-CHEMICAL COMBUSTION ANALYSIS

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Application October 2, 1952, Serial No. 312,835

6 Claims. (Cl. 23—230)

This invention relates to a method of sample burning and more particularly to a method of sample burning for micro-chemical combustion analysis of a sample contained in a combustion tube.

In making a micro-chemical combustion analysis it is common laboratory practice to place the sample to be analyzed in a glass or quartz combustion tube. This tube is generally comprised of four sections, a sample burning section which is the immediate area in the tube about the sample, a preheating section at the entrance end portion of the tube adjacent one end of the sample section, a sweeping section adjacent the other end of the sample section, and a section at the exit end of the tube adjacent the other end of the sweeping section which contains a catalyzing material to insure complete conversion of carbon to carbon dioxide. The tube containing the sample to be analyzed is rapidly heated to a temperature of from 800° C. to 1200° C. or higher, sufficient to burn the sample. During the period of heating when sample vapors are generated in the tube, oxygen is caused to flow through it downstream from the entrance end through the preheating, sample and sweeping sections respectively and into the heated catalyst section. The sample vapors combine with this oxygen to form gaseous oxides which are carried through the tube by the flow of oxygen and absorbed in absorbing reagents contained in separate vessels which are connected to the exit end of the combustion tube. These vessels are weighed before and after the absorption of the sample vapors.

In making micro-chemical combustion analyses, a number of problems arise, particularly when conducting large volumes of this work. Under such conditions it is necessary to have a standardized heating program technique which efficiently burns the sample, permits substantially all of the generated sample vapors to move into the catalyst section, and gives comparable results. It is therefore important to prevent small losses during sample burning because of the large error produced by such small losses in micro-chemical analysis. To accomplish this, diffusion of the generated sample vapors upstream in the combustion tube toward the preheating section from the sample section should be prevented and in burning the sample condensation or absorption of the generated sample vapors in the preheating section of the combustion tube should be kept at a minimum.

I have discovered a method of sample burning which overcomes these problems. Essentially, the preheating and sample burning sections of the combustion tube are heated to a predetermined temperature. However, the heating of the sample burning section is initiated after beginning heating of the preheating section and the temperature of the preheating section is maintained at a value greater than that of the sample burning section during generation of any of the sample vapors. A forward moving thermal-gradient is thus set up in the tube during generation of the sample vapors. As a result of this thermal-gradient, the generated sample vapors are prevented from diffusing upstream toward the preheating section. Instead, they are caused to move downstream toward and into the catalyst section with little or no loss due to back diffusion from the sample section into the preheating section or of condensation in the preheating and sample sections.

In applying my method, I prefer to employ a tube having a sweeping section located between the catalyst and sample burning section and to heat the catalyst section in order to accelerate oxidation reaction. Under such conditions, the sections are heated to predetermined temperatures. However, heating of the sample section is initiated after beginning heating of the preheating section; heating of the sweeping section is initiated after beginning heating of the sample section. During the period that sample vapors are being generated and until substantially all of these vapors have moved into the catalyst section, the temperature of the preheating section is maintained at a value greater than that of the sample section, which is maintained at a higher temperature than that of the sweeping section. In this way a forward moving thermal-gradient is set up in the combustion tube moving in the direction of the catalyst section and from the preheating, sample and sweeping sections respectively, until all of the sample vapors have moved into the catalyst section.

In most cases it is desirable to carry out the heating operation rapidly and during heating, causing oxygen to flow through the combustion tube from the entrance end toward the exist end to insure rapid and complete oxidation reaction of the sample vapors.

A radiant type heating furnace suitable for carrying out the process of this invention is illustrated in my copending application, Serial No. 307,681, filed September 3, 1952, now issued as Patent No. 2,671,123 dated March 2, 1954. Details of this apparatus suitable for carrying out the process of this invention are illustrated in the accompanying drawings, in which:

Figure 1 is a fragmentary, side elevational view of the furnace showing the combustion tube partly in dotted lines;

Figure 2 is an inside end elevational view of the furnace illustrating the movable half section of the furnace in two positions;

Figure 3 is a fragmentary, side elevational view of the furnace in an open position;

Figure 4 is a fragmentary sectional view taken along line 4—4 of Figure 1;

Figure 5 is a fragmentary sectional view taken along line 5—5 of Figure 4; and

Figure 6 is a fragmentary, diagrammatic view showing one form of positioning radiant heating furnaces of the type shown in Figures 1–5 so as to practice the method of this invention.

Inasmuch as the structural detail of the furnace is no part of this invention, reference is here made merely to the basic principles thereof.

As illustrated in Figures 1–5, a thin cylindrically shaped casing **10** is comprised of two inter-fitting semi-cylindrical half sections **11** and **12**, preferably made of aluminum. The inner surface of this casing is generally smooth and has an aluminum oxide coating thereon. A self-sustaining resistance wire heating element **13** is positioned within the confines of this casing and adapted to hold a quartz or glass combustion tube **14** therein, concentrically about and parallel to the longitudinal axis of the casing.

The casing **10** includes semi-circular shaped end plates **15** which, when the casing is closed, form two opposing circular end plates. Each of these have a hole therethrough to permit the combustion tube **14** to extend beyond the ends of the casing.

As shown in Figures 2, 3, and 4, one of the half

sections 11 of the casing is fixedly mounted at the bottom end portions atop the support elements 16. As here shown, the longitudinally extending edges of this section are about ten degrees from vertical. The other half section 12 is secured to the end portions of arms 17, each of the semi-circular end plates of this section being welded to one of these arms. The other end of each of these arms is rotatably mounted to upwardly extending posts 18 above the level of the top of the casing 10. By movement of the arms 17 upwardly, half section 12 is moved away from fixedly mounted half section 11, thereby opening the casing 10 and permitting lateral insertion and removal of the combustion tube 14. It is to be noted that opening of the casing 10 in the above described manner permits rapid cooling of a sample contained in the combustion tube and heated in the casing 10.

Self-sustaining resistance wire heating element 13, as shown in Figures 3, 4 and 5, consists of a plurality of spaced, generally parallel U-shaped self-sustaining wire heating segments 20 which are joined together at alternate ends of the U to form a continuous, longitudinally extending open-sided heating element. This heating element is mounted in the casing 10 by means of resistance wire leads 21 which extend downwardly from the heating element at the ends thereof through passageways 22 in the fixedly mounted half section 11 and into the support elements 16 where they are electrically adapted to be connected to an electrical source of power not shown in the drawings. This open-sided heating element 13 is adapted to hold the combustion tube 14 between the legs of the U-shaped segments 20 and is positioned within the casing 10 so that the combustion tube, when mounted between the legs of the U-shaped segments, is concentric about and extends parallel to the longitudinal axis of the casing 10. In addition, the open side of the heating element 13 faces the movable half section 12 in such a manner that a plane passing through the ends of the U-shaped segments 20 is substantially perpendicular to the horizontal plane passing through the longitudinal axis of the casing 10. In this way, when the half section 12 is moved away from the fixedly mounted half section 11 and to the position as shown in dotted lines in Figure 2, the combustion tube 14 can be inserted laterally between the legs of the U-shaped segments 20 or removed laterally therefrom.

An example of my new method employing the type of furnace described above is given below. As shown diagrammatically in Figure 6, the glass or quartz combustion tube 14 may extend through four adjacent radiant type heating furnaces. As here shown, this tube is divided into four adjacent sections in the following sequence: a preheating section 27 at the entrance end portion of the tube 14, a sample burning section 28 adjacent one end of the preheating section 27 and which contains a sample 29 that is to be burned for micro-chemical combustion analysis, a sweeping section 30 adjacent the other end of the sample section 28, and finally a section 31 adjacent the other end of the preheating section containing catalyzing material 32 to aid in accelerating oxidation of the sample vapors generated during burning of the sample. Each of the combustion tube sections 27, 28, 30 and 31 are enclosed by a separate stationary furnace 23, 24, 25 and 26, respectively.

In practicing my method, the resistance wire heating element 20*a* of the furnace 23 enclosing the preheating section 27 is energized through its leads 21*a* by an electrical source not shown in the drawings to cause heating of this section. After energizing the heating element 20*a*, the heating element 20*b* of furnace 24 enclosing the burning section 28 is energized to the point that permits burning of the sample 29. Following energization of the heating element 20*b*, the heating element 20*c* of furnace 25 is then energized. The electrical power input to each of the separate heating elements is controlled so as to heat the sections of the combustion tube 14 to predetermined temperatures, but during the period in which sample vapors are generated from the sample 29, the temperature of the preheating section is maintained at a value greater than that of the temperature of the burning sample section, which is maintained at a temperature higher than that of the sweeping section. In this way, a forward moving thermal-gradient is maintained in the combustion tube during generation of any sample vapors and until substantially all of the sample vapors have moved into the catalyst section. Consequently, diffusion of any sample vapors toward the preheating section is prevented and repeated uniform results can be obtained when conducting this work on a large volume basis.

While sample vapors are being generated in the combustion tube, the heating element 20*d* of furnace 26 may be energized and the electrical input to this furnace controlled so that the catalyst material is kept at a selected temperature without change or interruption. In this way the oxidation reaction is accelerated.

During heating of the combustion tube it is preferred to cause a stream of oxygen to flow through the tube in the direction of and into the catalyst section 31 from the preheating section 27 and through the sample burning and sweeping sections. As the oxygen passes through the tube, the various combustible elements of the sample combine with the oxygen to form gaseous oxides. The catalyzing material through which the gaseous oxides are carried insure complete conversion of carbon to carbon dioxide by accelerating the oxidation reaction.

After passing into the catalyst section, the gaseous oxides are caused to flow into separate vessels which contain absorbing reagents. These vessels, which are not shown in the drawings, are weighed before and after the absorption and the difference in weight determines the amount of gaseous oxides absorbed.

In the above heating procedure, each section of the tube once heated up is held at a temperature higher than the next forward section during the time in which the sample burning section is generating sample burning vapors, and the forward moving thermal-gradient resulting therefrom is maintained until substantially all of the sample vapors have moved into the catalyst section.

It is to be noted that in practicing my new method of sample burning, the furnaces employed can be stationary and when operated in the above described manner, a faster operation and shorter over-all time requirement per determination is achieved.

My method of heating the combustion tube may be accomplished by employing any number of separate furnaces or by employing a single furnace having adjacent multiple heating elements within the casing of the furnace, each element serving to heat a particular section and having separate electrical power input leads for so heating.

Instead of having a number of adjacent multiple heating elements within the casing, the furnace may contain a single long element with electrical power input taps to permit sectional step-wise energizing.

The foregoing detailed description is given for clearness of understanding only, and no unnecessary limitations should be understood therefrom, for some modifications will be obvious to those skilled in the art.

I claim:

1. The method of sample burning for micro-chemical combustion analysis of a sample contained in a combustion tube having a sample burning section and a preheating section adjacent one end of the sample section which comprises the steps of: heating the preheating section to a predetermined temperature; thereafter heating the sample heating section to a predetermined temperature lower than that of the preheating section; and maintaining the temperature of the preheating section higher than that of the sample burning section during the period that the sample burning section is generating sample vapors and thus producing a forward moving thermal-gradient in the

combustion tube in the direction toward the sample burning section from the preheating section and preventing diffusion of the sample vapors rearwardly toward the preheating section.

2. The method of sample burning for micro-chemical combustion analysis of a sample contained in a combustion tube having a sample burning section, a preheating section adjacent one end of the sample section, and a section containing a catalyst adjacent the other end of the sample burning section which comprises the steps of: heating the preheating section to a predetermined temperature; thereafter heating the sample burning section to a predetermined temperature lower than that of the preheating section; maintaining the temperature of the preheating section higher than that of the sample burning section during generation of any sample vapors in the sample burning section and until substantially all of the sample vapors have moved into the catalyst section thus producing a forward moving thermal-gradient in the combustion tube in the direction toward the catalyst section from the preheating and sample burning sections respectively; heating the catalyst section to a predetermined temperature lower than that of the sample burning section; and maintaining the temperature of the catalyst section lower than that of the sample burning section during generation of any sample vapors in the sample burning section until substantially all of the sample vapors have been moved into the catalyst section.

3. The method as set forth in claim 2 in which oxygen is caused to flow through the combustion tube toward the catalyst section from the preheating and sample burning sections during heating of the sections and until substantially all of the sample vapors move into the catalyst section.

4. The method of sample burning for micro-chemical analysis of a sample contained in a combustion tube having a sample burning section, a preheating section adjacent one end of the burning section, and a sweeper section adjacent the other end of the sample burning section which comprises the steps of: heating the preheating section to a predetermined temperature; thereafter heating the sample burning section to a predetermined temperature lower than the temperature of the preheating section; then heating the sweeping section to a predetermined temperature lower than that of the sample burning section; and maintaining the temperature of the preheating section higher than that of the sample burning section and the temperature of the sample burning section higher than that of the sweeping section during the period that the sample burning section is generating sample vapors thus producing a forward moving thermal-gradient in the combustion tube in the direction toward the sweeping section from the preheating and sample burning section and preventing diffusion of sample vapors rearwardly toward the preheating section.

5. The method of sample burning for micro-chemical analysis of a sample contained in a combustion tube having a sample burning section, a preheating section adjacent one end of the sample section, a sweeping section adjacent the other end of the sample section, and a section containing a catalyst adjacent the other end of the sweeping section which comprises the steps of: heating the preheating section to a predetermined temperature; thereafter heating the sample burning section to a predetermined temperature lower than the temperature of the preheating section; then heating the sweeping section to a predetermined temperature lower than the temperature of the sample burning section; maintaining the temperature of the preheating section higher than the temperature of the sample burning section and the temperature of the sample burning section higher than the temperature of the sweeping section during generation of any sample vapors in the sample burning section thus producing a forward moving thermal-gradient in the combustion tube in the direction toward the catalyst section from the preheating and sample burning sections; heating the catalyst section to a predetermined temperature lower than the temperature of the sweeping section; and maintaining the temperature of the catalyst section lower than the temperature of the sweeping section during generation of any sample vapors in the sample burning section and until substantially all of the sample vapors have moved into the catalyst section.

6. The method as set forth in claim 5 in which oxygen is caused to flow through the combustion tube toward the catalyst section from the preheating and the sample burning sections during heating of the sections and until substantially all of the sample vapors have moved into the catalyst section.

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