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ANALYZER FOR DETERMINING HYDROGEN CONTENT OF GAS

Filed March 26, 1963

2 Sheets-Sheet 1

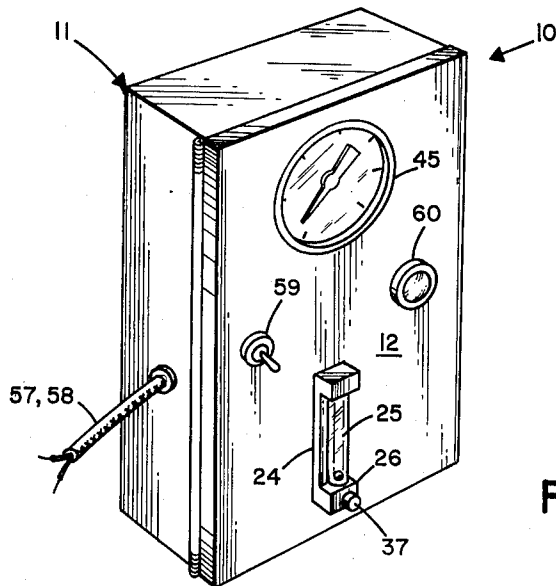


Fig. 1

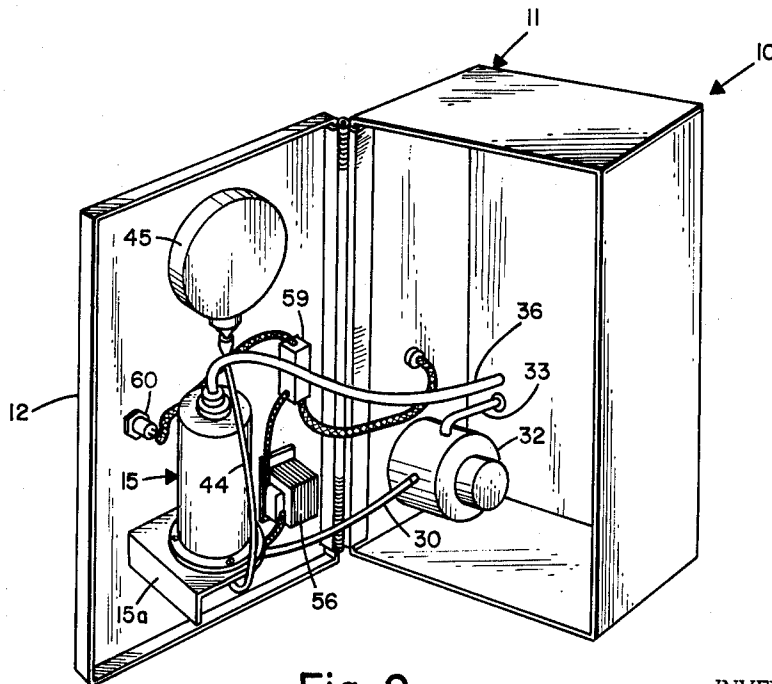


Fig. 2

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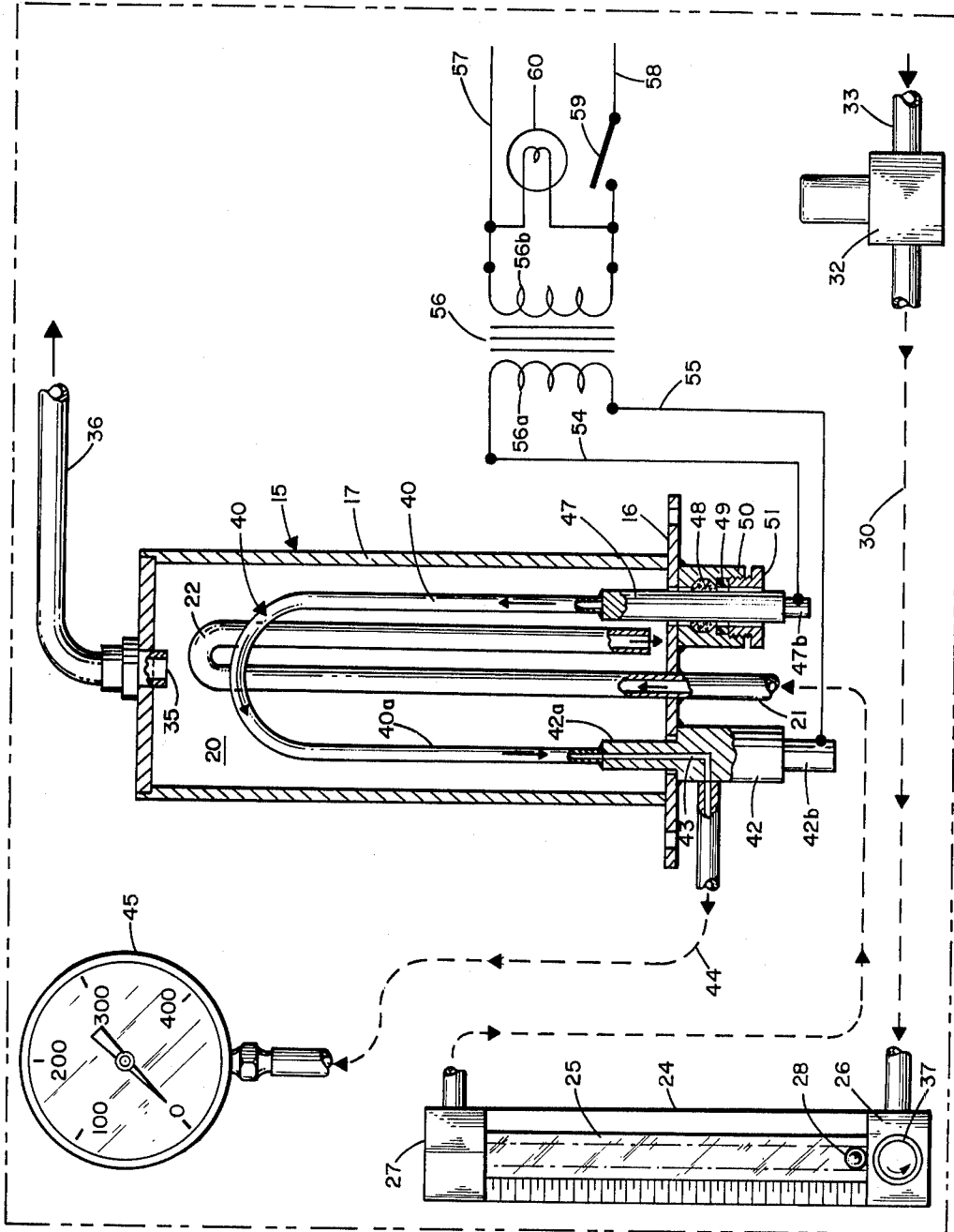


Fig. 3

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ANALYZER FOR DETERMINING HYDROGEN CONTENT OF GAS

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This invention relates to improvements in gas analyzers, and more particularly to an improved gas analyzer for determining the amount of hydrogen present in a gas constituting a mixture of hydrogen and other gasses.

The analyzer of this invention utilizes the well known principle that in a mixture of gasses under pressure, the total pressure exhibited by the gasses is equal to the sum of the pressures exerted by each of the gasses individually. This is known as the Law of Partial Pressures, the pressure exhibited by each of the gasses being referred to as its partial pressure. It is also a well known fact that hydrogen gas is alone in its capability of passing through palladium metal or certain palladium metal alloys such as palladium silver, whereby palladium alloys have been utilized as a filter material for purifying hydrogen. The ability of palladium to pass hydrogen gas is enhanced by maintaining the temperature of the palladium or palladium alloy in a range of 400° F. to 1000° F., and this has been accomplished heretofore through the use of separate heating elements, such as electrical resistance type heaters, mounted adjacent the palladium filter element.

It is a primary object of this invention to provide an improved gas analyzer utilizing the foregoing principles of partial pressure and hydrogen transmission by palladium alloys to determine the percent molar content of hydrogen in a gas of unknown hydrogen content, the device comprising a heated hollow palladium alloy element the exterior of which is subjected to a stream of hydrogen containing gas to be tested, the absolute pressure of which may be taken as the total of the partial pressures of the constituent gasses thereof. The interior of the palladium alloy element is substantially free of gasses other than hydrogen and is connected to a suitable pressure indicating means. Hydrogen will pass through the wall of the palladium alloy element in either direction until the pressure therein is substantially equal to the partial pressure of the hydrogen component of the gas being tested. The pressure within the palladium alloy element may be read from the indicating means, and the ratio between the absolute pressure within the element to the absolute pressure of the tested gas provides an accurate indication of the percent mole hydrogen in the gas tested.

As another object, this invention aims to provide a particularly simple, safe, yet reliable gas analyzer of the foregoing character wherein no separate heating means are required for heating the palladium alloy element to the preferred temperature range, the element being self heated by a flow of electric current therethrough which is converted to heat energy by the inherent electrical resistance of the element.

It is another object of this invention to provide a hydrogen gas analyzer utilizing a palladium alloy element heated by its own resistance to electric current flow, and which element comprises a palladium silver alloy tube housed in a chamber through which the gas to be analyzed flows at a predetermined rate under a predetermined pressure. In one practical embodiment of the invention, the palladium silver tube has a U-shaped configuration and is mounted in the test gas chamber by

securing the ends of the legs of the element to the base of the chamber, one leg being electrically insulated from the base so that an electrical potential established across the legs will result in a current flow and self heating of the element.

The invention may be further said to reside in certain novel combinations and arrangements of parts hereinafter described in conjunction with the accompanying sheets of drawings forming a part of this specification, and in which

FIG. 1 is a perspective view of a gas analyzer embodying this invention;

FIG. 2 is a perspective view of the gas analyzer of FIG. 1 with the front cover open to reveal the interior parts thereof; and

FIG. 3 is a diagrammatic illustration of the gas analyzer of this invention.

In the form of the invention shown in the drawings and described hereinafter, there is provided a gas analyzer 10 comprising a suitable rectangular cabinet 11 having a hinged front panel 12. Various components of the apparatus, described more fully hereinafter, are mounted on the front panel 12 or within the cabinet.

Referring to FIG. 3, the heart of the gas analyzer comprises a hydrogen diffusion cell 15 including a base plate 16, a cylindrical shell 17, and an end plate 18 welded together to define a gas tight chamber 20. The cell 15 is conveniently mounted on a bracket 15a on the inner surface of the hinged panel 12 as shown in FIG. 2. Gas is admitted into the chamber 20 through a tube 21 which extends through an opening in the base plate 16 and is formed into an inverted U-shaped preheater tube 22. The tube 21, which is soldered in its base plate opening, leads to the outlet side of a flow meter 24 mounted on the face of panel 12.

The flow meter 24, which may be of any type well known in the art, is shown here as being of the type comprising a glass tube 25 connected to define a gas passage between an inlet header 26 and an outlet header 27 containing a sphere 28 movable in the tube 25. The flow of gas through the tube 25 acts upwardly on the sphere 28 so that at a predetermined known flow rate the force of the fluid acting on the sphere equals the force of gravity and the sphere will "float" between the ends of the tube. The flow meter 24, which is conveniently secured to the front of the front panel 12 of cabinet 11, is connected by an inlet tube 30 to a pressure regulator 32 which is in turn connected by a line 33 to a source of gas to be analyzed containing an indeterminate percentage of hydrogen. The pressure regulator 32, which is conveniently mounted inside the cabinet 11, is adjustable to provide a substantially constant pressure in tube 30 irrespective of changes in the supply pressure.

The chamber 20 of diffusion cell 15 is provided with an outlet connection 35 and a tube 36 leading to atmosphere or to a suitable drain. The pressure regulator 32 and control valve 37 can be adjusted to produce a predetermined rate of flow through the cell as well as to maintain a predetermined pressure therein.

It will be seen that the chamber 20, flow meter 24, valve 37, and pressure regulator 32 are connected in series. When the pressure within the chamber 20 is to be 15 p.s.i.a. or less, the series connections are made with the elements in the order illustrated in the drawings and described above. However, when the pressure in chamber 20 is to be above 15 p.s.i.a., the order of series connection is advantageously changed so that the gas flow from pressure regulator 32 goes directly to the chamber 20 through the preheater tube 22, then through the valve

37 and flow meter 24 to drain line 36. This permits the use of valve 37 as a back up valve against which the desired pressure may be developed.

Mounted within the chamber 20 and forming part of the hydrogen diffusion cell is a U-shaped tubular element 40 formed of a palladium and silver alloy (Pd—Ag). The element 40 has one leg 40a silver soldered to the reduced end 42a of an outlet manifold 42 which extends upwardly through an opening in the base plate 16 and is silver soldered thereto in gas tight relation. The manifold 42 has a passageway 43 communicating with the interior of the element 40 and through a tube 44 with a pressure gauge 45 which in this example is a 0 to 400 p.s.i.g. gauge.

The other leg 40b of the element 40 is soldered to a support terminal member 47 which closes the end of leg 40b. The terminal member 47 extends through an opening in base plate 16 and through packing 48 and a packing washer 49 held in a packing gland 50. The packing gland 50 is welded to plate 16 and receives a gland nut 51 which is tightened to cause the packing to effect a gas tight seal about terminal member 47.

The palladium silver tubular element 40 is electrically insulated from the remainder of the cell 15 by the packing and is adapted to be subjected to a flow of electric current from one end thereof to the other to effect resistance heating thereof. To this end, the manifold 42 and terminal member 47 are provided with terminals 42b and 47b, connected by wires 54 and 55 to the secondary winding 56a of a step down transformer 56. The primary winding 56b of the transformer is supplied with electric current, such as 115 v. A.C. through line conductors 57, 58 and a suitable on-off switch 59 in conductor 58. A pilot light 60 is conveniently connected across conductors 57, 58 to indicate energization of the apparatus upon closing switch 59. Both the switch 59 and the pilot light 60 are conveniently mounted through the panel 12, as shown.

The tubular, U-shaped palladium silver (Pd—Ag) element will be subjected to an alternating current flow therein, which, because of the inherent electrical resistance of the element, will result in heating thereof. In the present example, the transformer provides a voltage across its secondary, and hence between the ends of the element 40, of 2.5 volts, and the element 40 comprises an eight inch length of one eighth inch outside diameter Pd—Ag tubing with .005 inch wall thickness. These dimensions of element 40, when a voltage of 2.5 volts is applied thereacross and gas to be tested is flowing through the cell 15 at a rate of about 2.5 cubic feet per hour, will produce a temperature of the element in the desired range of 400° F. to 1000° F.

In the operation of the apparatus shown, the interior of the element 40 is evacuated and purged with pure hydrogen since the presence of gas other than hydrogen will introduce an error in the results. The switch 59 is closed to commence the self heating of the U-shaped element 40 in the cell 15, and the pressure regulator 32 is connected to a source of gas to be tested and set to maintain in the cell 15 a predetermined constant pressure selected within a range of less than 15 p.s.i.a. with the control valve 37 set to maintain a flow of 2.5 cubic feet per hour indicated at the flow meter 24. It will be understood that when higher pressures in the range of 15 p.s.i.a. to 400 p.s.i.a. are used, the series connections of the element of the apparatus will be arranged as described above. In this event, the adjustments are substantially the same. That is, the pressure regulator 32 and valve 37 are manipulated to provide the desired pressure and rate of flow.

The preheater tube 22 is disposed adjacent the electrically heated palladium-silver element and is heated thereby so that as the sample gas flows therethrough it is in turn heated. Hydrogen in the preheated sample gas will then readily diffuse through the heated walls of the palladium-silver element 40 until the pressure of the

hydrogen within the element is substantially equal to the partial pressure exerted by hydrogen in the sample gas on the outside of the element 40. The time required for the hydrogen pressures inside and outside the element 40 to equalize as indicated by stabilization of the reading of gauge 45, varies with the total pressure of the sample gas, and in actual practice has been found to require from one-half to eight minutes, with the longer times being experienced where the higher operating pressures are used.

When the gauge indication is stabilized the reading thereof is taken and the molar concentration of hydrogen in the sample is readily calculated as the ratio of the H₂ partial pressure (absolute) to the total pressure (absolute). Assuming the total pressure to be held constant with accuracy by the regulating valve 32, the gauge 45 can be calibrated directly in the mole percent H₂.

By way of example, if a ternary gas consisting of 50% H₂, 25% N₂ and 25% Xe, is used as a gas to be tested, under a pressure of 200 p.s.i.a. the H₂ would have a partial pressure of 100 p.s.i.a., the N₂ would have a pressure of 50 p.s.i.a., and the Xe would have 50 p.s.i.a. Since only the hydrogen will pass through a heated Pd—Ag tube, if the cell 15 is provided with a flow of ternary gas at a total pressure of 200 p.s.i.a. surrounding the tube 40, the gauge will stabilize at a reading of 100 p.s.i.a. Then, by the above mentioned ratio

$$\frac{100 \text{ p.s.i.a.}}{200 \text{ p.s.i.a.}} = 50\%$$

Thus, the ternary gas is found to have a 50% H₂ content.

Of course, the transformer 56 could be replaced by a D.C. power supply such as batteries or regulated rectifier means. Also, the gauge 45 may be replaced by a pressure transducer, the electrical output of which is applied to a suitable recorder for providing permanent, direct, and continuous readings of the H₂ content of the sampled gas.

It will be recognized that as readings move downscale, the hydrogen will actually be passing from within the tubular element 40 into the sample gas stream, until the pressure in the tubular element equals the partial pressure in the sample. Conversely, as the readings move upscale, hydrogen will be passing from the sample gas stream into the tubular element 40.

The apparatus may be accurately calibrated by operation with prepared sample gasses having known hydrogen contents.

Although the invention has been described in considerable detail with reference to a specific gas analyzer apparatus embodying the invention, it will be understood that the invention is not limited thereto, but rather the invention includes all those modifications, adaptations, substitutions, changes, and uses as are reasonably embraced by the scope of the claims hereof.

Having described my invention, I claim:

1. Gas analyzing apparatus of the character described comprising:

- (a) a body defining a chamber having an inlet and an outlet for a gas to be analyzed,
- (b) conduit means from a source of gas to said inlet and including a pressure regulating valve connected therein between said source of said gas and said inlet,
- (c) a flow meter connected in said conduit means to indicate the rate of gas flow therethrough,
- (d) a tubular palladium-silver element disposed in said chamber, one end of said element being closed,
- (e) pressure indicating means communicating with the interior of said element through the other end thereof,
- (f) means electrically insulating at least one end of said element from said chamber defining body, and
- (g) means connected to the opposite end of said element for causing an electric current to flow therethrough, whereby said element is heated in accordance with its electrical resistance, said conduit means including an appreciable length of tubing in said chamber subjected to the heat of said element for

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preheating gas discharged therefrom into said chamber.

2. Gas analyzing apparatus of the character described comprising:

- (a) a body defining a chamber having an inlet and an outlet for a gas to be analyzed, 5
- (b) conduit means from a source of gas to said inlet and including a pressure regulating valve connected therein between said source of said gas and said inlet, 10
- (c) a flow meter connected in said conduit means to indicate the rate of gas flow therethrough,
- (d) a tubular palladium-silver element disposed in said chamber, one end of said element being closed,
- (e) pressure indicating means communicating with the interior of said element through the other end thereof, 15
- (f) means to heat said palladium alloy element, and
- (g) said conduit means including an appreciable length of tubing in said chamber subjected to the heat of

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said element for preheating gas discharged therefrom into said chamber.

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