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RADIOLOGICAL GAS ANALYSIS
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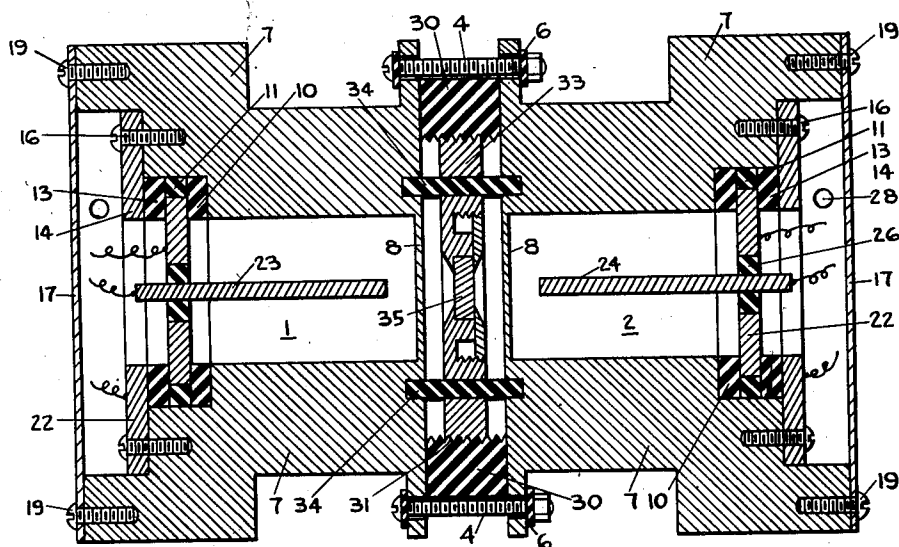


Fig. 1

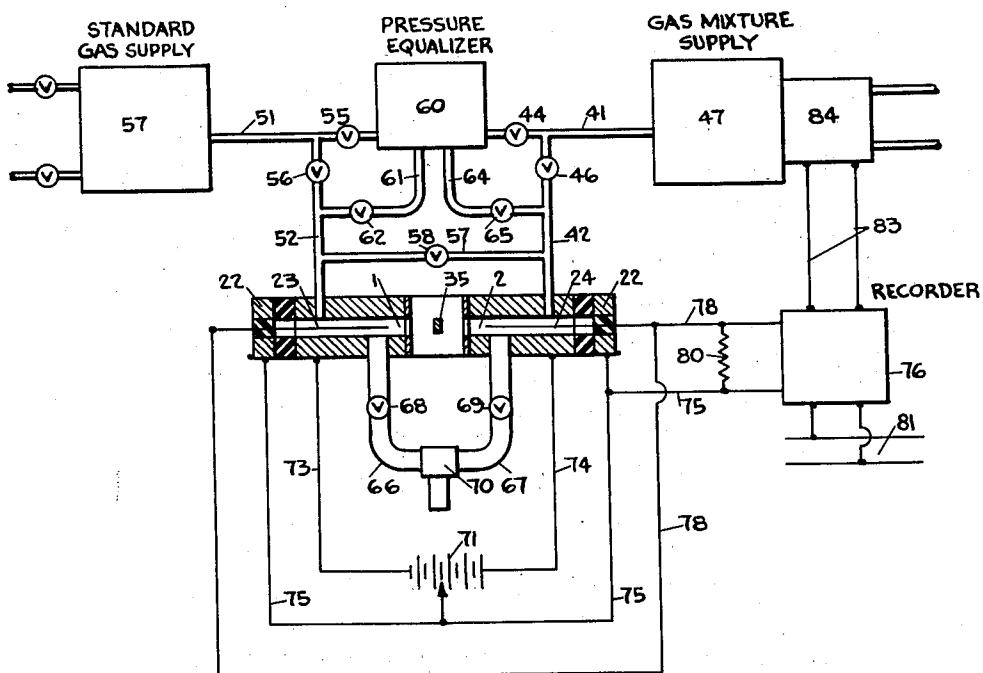


Fig. 2

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RADIOLOGICAL GAS ANALYSIS

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This invention relates to gas analysis, and pertains more particularly to radiological methods and apparatus for analyzing gaseous mixtures.

It is known that radioactive emanations or radiations, such as alpha, beta and gamma rays, and under certain conditions radiations of the neutron and other types, may be detected by the ionizing effects which they produce in passing through a space forming the field between a pair of suitably charged electrodes. The radiations, passing through said space, cause the production of ion pairs, of which the positively charged ions travel to the negative electrode, and the negatively charged ions or electrons travel to the positive electrode.

The various radiation-detecting devices operating on this principle may be generally defined as ion chambers, and are more particularly classified as Geiger counters, proportional counters and ionization chambers, depending chiefly on the potential applied between the electrodes and the pressure of the gaseous medium surrounding said electrodes.

The Geiger counters operate normally at relatively high voltages, the proportional counters at intermediate voltages, and the ionization chambers at relatively low voltages. While the total operating voltage range of ion chambers may thus extend from a few tens of volts to several thousand volts, the demarcation lines between the particular voltage ranges of the Geiger counters, proportional counters and ionization chambers are by no means rigidly set and often overlap depending on other operating conditions, such as the pressure of the gaseous medium between the electrodes. Normally, sub-atmospheric pressures are used with Geiger and proportional counters, while atmospheric or super-atmospheric pressures up to several thousand pounds per square inch are used with ionization chambers.

It has been found according to the present invention that gas mixtures, and particularly binary gas mixtures, may be accurately analyzed for the proportions of their component gases by taking advantage of the difference in the ionization cross-section of said component gases.

The ionization cross-section of a gas, which may be defined as the effective area of a molecule of said gas presented to the impact of an ionizing particle, is a quantity determined by the size of a gas molecule and the number of electrons in the atoms forming said molecule. The

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ionization cross-section determines the number of ions produced per ionizing particle under predetermined pressure and temperature conditions. An ion chamber filled with a gas mixture and exposed to an external source of atomic or subatomic particles or radiation will therefore produce in a circuit connected between its electrodes, a current whose intensity is a function of the composition and the total pressure of the gas in the ion chamber.

It is, therefore, an object of this invention to provide a radiological method of gas analysis, and an apparatus for carrying out such analysis.

It is also an object of this invention to provide for this purpose an improved ionization chamber especially suitable for the analysis of binary gas mixtures.

These and other objects of this invention, which comprises the control of the compositions of the gases being analyzed, will be understood from the following description taken with reference to the appended drawings, wherein:

Fig. 1 is a cross-section view of the analytical unit of the present invention, said unit comprising two identical ion chambers and a radioactive source.

Fig. 2 diagrammatically shows the unit of Fig. 1 connected into the electrical and gas pressure system of the present invention.

Referring to the drawings, it is expressly understood that any type of ion chamber, as defined hereinabove, may be used for the purposes of this invention, depending on the type of analysis which is to be carried out, and the range of voltages and pressures which it is desired to utilize. To simplify exposition, however, the invention will be described with regard to a particular ionization chamber structure, it being further understood that the use of such ionization chamber forms a preferred embodiment of the invention in view of the special advantages to be derived therefrom, as will appear hereinbelow.

Referring to Fig. 1, the ion chamber unit of the present invention comprises two ionization chambers 1 and 2, preferably of a generally cylindrical shape, held in co-axial adjustment with each other by means such as a plurality of circumferentially disposed screws 4 provided with insulating gaskets 6. Chambers 1 and 2 are preferably substantially identical.

The walls 7 of the chambers 1 and 2 are made of a suitable material, such as aluminum, magnesium, copper, bronze, brass and their alloys, etc. Those portions of the walls which close the

coaxial inner ends of chambers 1 and 2 have their thickness greatly reduced (to a value such, for example, as 0.01 of an inch) to facilitate the penetration of radiation into said chambers. The use of light metals such as magnesium or aluminum in at least these portions of the walls is preferred because of the permeability of these metals to weak or soft radiation.

The outer ends of the cylindrical chambers 1 and 2 have their inside diameter enlarged in a stepwise manner to accommodate annular gas-kets 10, centering rings 11, insulator rings 13 and retainer rings 14, held to the chamber walls 7 by means such as screws 16. The centering and insulating rings and the gaskets are made of a suitable plastic material such as hard rubber, plastic, glass, etc., while the retainer rings are preferably of the same metal as the chambers. The outer ends of chambers 1 and 2 are closed by means of metallic back cover plates 17 held to the walls 7 by means such as screws 19.

Held between the insulating annular elements 10, 11 and 13 are metallic guard rings 22 supporting electrodes 23 and 24 by means of insulator bushings 26, made of hard rubber, plastic, glass, etc. Windows 28 are provided in the chamber walls 7 or cover plates 17 to permit suitable electrical connections to be made to the electrodes and guard rings. Mounted between the chambers 1 and 2 coaxially therewith is a rotatable adjusting ring 30 made of an insulating material as above, and having its inner wall screw-threaded, as shown at 31. An externally screw-threaded source holder ring generally designated by 33 is mounted within the adjusting ring in screw-threaded engagement therewith. Guide rods 34, likewise made of an insulating material, and removably affixed to the walls of chambers 1 and 2, slidably pass through the body of the holder 33 and prevent it from rotating. Rotation of the adjusting ring 30 thus results in an axial displacement of the holder 33 to the left or right. The amount of ionizing radiation received by chambers 1 and 2, respectively, from a radioactive source 35, suitably mounted in the holder 33, can thus be equalized or adjusted to regulate the output currents of said chambers by rotating the adjusting ring in the proper direction.

Chambers 1 and 2 are electrically insulated from each other by means of the gaskets 6, adjusting ring 31 and guide rods 34.

Any suitable radioactive element giving rays of a suitable type such as alpha, beta or gamma, may be used for the purposes of this invention. It is, however, preferred to use a source of beta rays, since these rays have the advantage over gamma rays of a high ionizing power, thus giving fairly large ionization currents, and of a relatively low penetration power, thus making it unnecessary to use heavy shielding. The beta rays also have the advantage over alpha rays of a sufficiently high penetration power to permit the use of moderately thick windows (for example, 0.01" of aluminum) in the radiation detector, which permits a more rugged construction and facilitates the use of two detectors in opposition for differential measurements. The source may consist of radioactive carbon C¹⁴, or of a suitable fission product, such as strontium 90, which is especially desirable since it delivers beta radiation of a high energy of about 2 Mev., and has

a half-life of about 25 years. Possible sources may further be selected from the table below:
Radioactive isotopes

Isotope	Half-life	Emission		
		Alpha	Beta	Gamma
Strontium 90.....	30 years.....	No	Yes	No
Yttrium 90.....				
Cerium 144.....				
Praseodymium 144.....	275 days.....	No	Yes	Yes
Ruthenium 106.....				
Rhodium 106.....	1 year.....	No	Yes	Yes
Carbon 14.....	6,000 years.....	No	Yes	No
Radium 226.....	1,600 years.....	Yes	Yes	Yes
Silver 110.....	225 days.....	No	Yes	Yes
Thallium 204.....	3 years.....	No	Yes	No

The isotopes listed above have reasonable half-lives and are readily available. The first three examples on the list are pairs in a decay chain. The first isotope in each pair has the half-life indicated; the second has a very short half-life and is in secular equilibrium with the parent isotope. The second isotope in each case emits the high energy beta particles of interest.

Fig. 2 shows diagrammatically the device of Fig. 1 connected into suitable electrical and gas supply circuits, in accordance with this invention.

A supply of a gas mixture which it is desired to analyze for the percent of its components, such for example as a mixture of argon and nitrogen, oxygen and nitrogen, carbon dioxide and air, propane and propylene, etc., is delivered to the chamber 2 from a reservoir or any other source of supply 47 through conduits 41 and 42 provided with valves 44 and 46. A sample of a standard gas, or of a standard gas mixture of known composition is delivered to the chamber 1 through pipes 51 and 52 provided with valves 55 and 56 from a source of supply 57. Thus, for example, if the reservoir 47 contains a mixture of propane and propylene of unknown proportions, the reservoir 57 may contain pure propane or pure propylene, or a mixture of accurately known proportions of propane and propylene.

Chambers 1 and 2 are provided with outlet pipes 66 and 67, having valves 68 and 69, which may be manifolded as at 70 to exhaust the contents of chambers 1 and 2 to any suitable zone of low pressure, such for example as the atmosphere. A pipe 57 provided with a valve 59 is connected between pipes 42 and 52.

Since, in order to eliminate total pressure effects, the gases in chambers 1 and 2 must be at equal and constant pressures, a pressure equalizer 60 of any desired conventional type may be connected between valves 44 and 55 to ensure that the gases delivered to chambers 1 and 2 from reservoirs 47 and 57, respectively, are at the same constant pressure. The pressure equalizer is connected to chamber 1 by a conduit 61 provided with a valve 62, and to the chamber 2 by a conduit 64 provided with a valve 65.

However, a simpler and preferred method of equalizing pressures in chambers 1 and 2 may be used, which consists in providing exhaust pipes 66 and 67 of substantially larger diameter, and therefore throughput capacity, than the supply pipes 42 and 52. In such case the pressure of the gases from reservoirs 47 and 57, upon admission to chambers 1 and 2, drops substantially instantaneously to the pressure value of the exhaust zone, such as the atmosphere, and the pressure of the chambers is thus suitably equalized and maintained constant for the purposes of this invention. In order to operate the system of Fig. 2

in this manner, it is only necessary to open valves 46, 56, 68 and 69 and to close all the other valves.

The electrical circuits of the present system comprise a source of D. C. potential, which is shown in Fig. 2 as a battery 71, but which can obviously consist of any other source, such for example as a D. C. generator or a rectifier provided with a suitable filter. One of the terminals of source 71 is connected to the wall of chamber 1 and the other to the wall of chamber 2 by leads 73 and 74, while the mid-voltage point of source 71 is connected in parallel by lead 75 to the two guard rings 22 of chambers 1 and 2, said guard rings being thus at ground potential with regard to the source 71. Lead 75 is also connected to one of the terminals of a recorder 76, whose other terminal is connected in to the electrodes 23 and 24 in parallel. A resistor 80 at very high resistance, such as 10^{10} or 10^{12} ohms, is connected across the leads 75 and 78.

It will be seen that with this arrangement one of the chambers, for example chamber 1, will serve as a cathode with regard to the electrode 23 therein, while the other chamber 2 will serve as an anode with regard to the electrode 24.

The electrodes 23 and 24 are thus connected in opposition to each other, and the resulting differential current, which is a measure of the difference of ionization intensity in the chambers 1 and 2, may be applied directly to the recorder 76 without need for any intermediary balancing or measuring circuits.

The electrodes 23 and 24 have a relatively large diameter, such as about $\frac{1}{4}$ inch, to prevent the formation of a high gradient field such as occurs in the immediate vicinity of thin wire-type electrodes. In such high gradient field, undesirable secondary ionization may be produced by negative ions accelerated at an excessive rate toward the positive electrode, which would unbalance the chambers. The reason for grounding the guard rings 22 is to eliminate or minimize the effects of any leakage across insulators 10, 11, 13 and 26 of Fig. 1, by preventing any leakage current flow through the resistor 80. With the arrangement shown in Fig. 2, it will be seen that the guard rings and the electrodes are at substantially the same potential and any potential drop between the electrodes and the guard rings is therefore only that due to the flow of the ionization current through the resistor 80, said drop having a value of a few millivolts.

The recorder 76, which may be energized from any desired source, such as a power line 81, comprises amplifier and other circuits necessary for suitably amplifying, measuring and recording the voltage input supplied thereto across the resistor 80, in a conventional manner well understood in the art and forming no part of this invention. The output current of the recorder may be applied through leads 83 to operate an electrically controlled valve 84 or other agency capable of changing the composition of the gas in the reservoir 47, whereby said composition may be suitably controlled in accordance with industrial production requirements.

In describing the method of operation of the present system, it will be assumed that it is not desired to use the pressure equalizer 60, which will at all times remain cut out from the system by closing valves 44, 55, 62 and 65. Assuming that reservoir 47 contains a mixture of propane and propylene in unknown proportions, the electrical circuits of the system are energized and valve 46 to reservoir 47 is closed. Both chambers are flushed with pure propane from reservoir 57

through pipes 51, 52, 57 and 42, said propane being at atmospheric pressure in both chambers and being exhausted through manifold 70. In view of the opposite polarity of electrodes 23 and 24, the ionization currents delivered thereby are in opposition to each other, and whatever unbalance current is indicated at this time by the recorder can be readily balanced out by a suitable adjustment of the electrical circuits of the recorder, or by properly shifting the position of the radioactive source 35 by means of the ring 31, or both, until the recorder has a zero reading.

With propane from tank 57 flowing through chambers 1 and 2, valve 58 is closed which allows propane to flow only through chamber 1. The contents of reservoir 47 are then successively changed, as for example to 1, 2, 5, 10, 20 etc., percent propylene, and valve 46 is opened. Chamber 2 is then flushed successively with the above mixture and the recorder readings are taken each time to obtain a calibration curve, or to calibrate the recorder directly.

The calibrating operation completed, reservoir 47 is filled with the unknown mixture, valve 58 is closed and chamber 2 is flushed with the unknown mixture of propane and propylene from reservoir 47 through pipes 41 and 42. Chamber 1 is simultaneously flushed with propane and the percentages of the unknown propane-propylene mixture are obtained either directly from the calibrated recorder or by reference to the calibration curve. It is understood that the above procedure is given here merely by way of illustration, and that many variations thereof are possible without departing from the spirit of the invention, as will be understood by those skilled in the art.

The invention claimed is:

1. A method for analyzing a gaseous medium for its composition, comprising the steps of establishing two electrical fields of substantially equal constant potential in two separate zones, flowing a gas of known composition through each of said zones while maintaining the gas therein at substantially equal constant pressures, subjecting the gas in said two zones to beta radiation of substantially equal constant intensity, combining the ionization currents produced in said two zones by said radiation in opposition to each other in such a manner as to reduce the resultant current to zero, discontinuing the flow of the gas of known composition through one of the chambers, flowing a gas of unknown composition through said chamber while maintaining constant conditions of pressure and radiation substantially as before, combining the ionization currents produced in the two chambers in opposition to each other, and determining the composition of the unknown gas by measuring the resultant current.

2. Apparatus for analyzing a gaseous medium, comprising two ion chambers each having two electrodes, said chambers and said electrodes being disposed longitudinally of each other along a single axis, first conduit means opening to said chambers for supplying a gas of known composition to one of said chambers and a gas of unknown composition to the other chamber, means comprising second conduit means in gaseous communication with said first conduit means for equalizing and maintaining constant the pressure in said two chambers, circuit means for applying substantially equal constant potentials between the electrodes of each chamber, radioactive source means for subjecting said chambers to an ionizing radiation, said source being mounted be-

tween said chambers along the axis thereof, and electrical circuit means connected to one of the electrodes of each of said chambers for comparing the ionization currents delivered by said electrodes under the effect of said radiation.

3. Apparatus for analyzing a gaseous medium, comprising two substantially identical ion chambers each having two electrodes, first and second inlet conduit means for supplying a gas of known composition to one of said chambers and a gas of unknown composition to the other chamber, respectively, first and second outlet conduit means for exhausting the gases from said first and second chambers to a common low pressure zone, the throughput capacity of said outlet means being greater than that of said inlet means, circuit means for applying substantially equal constant potentials between the electrodes in each chamber, radioactive source means for subjecting said chambers to an ionizing radiation, and electrical circuit means connected to one of the electrodes of each of said chambers for comparing the ionization currents delivered by said electrodes under the effect of said radiation.

4. An analytical unit for use in combination with a gas analysis system comprising a supply of gas of known composition, a supply of gas of unknown composition, a source of electric power and an electrical measuring device, said unit comprising two ionization chambers, a source of ionizing radiation disposed between said chambers, inlet means in the first and the second chambers for admitting thereto a known and an unknown gas respectively, outlet means in the first and the second chamber for exhausting the gases therein to a common low pressure zone, the capacity of said outlet means being larger than the capacity of said inlet means, the walls of each of said chambers forming the outer electrode thereof, an inner electrode in each chamber, a guard holder in each chamber fixedly mounting the inner electrode with regard to the outer electrode, insulating means in each chamber between said guard holder and said inner and said outer electrodes, means for applying a potential between the outer electrodes of said two chambers, means for maintaining each guard holder at substantially the potential of the electrode mounted thereon, means for connecting the inner electrodes in opposition to each other, and means for measuring the resultant ionization current obtained from said inner electrodes.

5. An analytical unit for use in combination with a gas analysis system comprising a supply of gas of known composition, a supply of gas of unknown composition, a source of electric power and an electrical measuring device, said unit comprising two cylindrical ionization chambers arranged coaxially with regard to each other and electrically insulated from each other, a source of ionizing radiation mounted between said chambers on the axial line thereof, means for adjusting the position of said radiation source with regard to said chambers along said axial line, the thickness of the walls of each chamber adjacent said radiation source being greatly reduced, inlet means through the walls of each chamber for admitting a gas thereto, outlet means through the walls of each chamber for exhausting a gas therefrom, the diameter of said outlet means being larger than that of said inlet means, an inner electrode disposed in each chamber axially there-

of, a guard holder in each chamber supporting the inner electrode therein, said guard holder being electrically insulated from the electrode and from the chamber walls, means for applying an electric potential between each chamber and the electrode therein, and means for maintaining each guard holder substantially at the potential of the electrode supported thereby.

6. The apparatus of claim 2, wherein the means for subjecting the gas to an ionizing radiation comprises essentially a source of beta rays.

7. The apparatus of claim 2, wherein the means for subjecting the gas to an ionizing radiation is strontium 90.

8. The apparatus of claim 2, wherein the inner electrode is of a substantial thickness of not less than 0.1 inch diameter.

9. An analytical unit for use in combination with a gas analysis system comprising gas supply means, a source of electric power, and an electrical measuring device, said unit comprising two cylindrical ion chambers arranged coaxially with regard to each other, a source of ionizing radiation mounted between said chambers on the axial line thereof, means for adjusting the position of said radiation source with regard to said chambers along said axial line, inlet means through the walls of each chamber for admitting a gas thereto, outlet means through the walls of each chamber for exhausting a gas therefrom, inner and outer electrode means in each chamber, and means for applying an electric potential between the electrodes in each chamber.

10. Apparatus for analyzing a gaseous medium, comprising two substantially identical ion chambers each having two electrodes, first and second inlet conduit means for supplying a gas of known composition to one of said chambers and a gas of unknown composition to the other chamber, respectively, first and second outlet conduit means for exhausting the gases from said first and second chambers to a common low pressure zone, the throughput capacity of said outlet means being sufficient to maintain said chambers substantially at the pressure of said low pressure zone, circuit means for applying substantially equal constant potentials between the electrodes in each chamber, radioactive source means for subjecting said chambers to an ionizing radiation, and electrical circuit means connected to one of the electrodes of each of said chambers for comparing the ionization currents delivered by said electrodes under the effect of said radiation.

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References Cited in the file of this patent

UNITED STATES PATENTS

Number	Name	Date
2,097,760	Failla	Nov. 2, 1937
2,437,476	Parker	Mar. 9, 1948
2,495,650	Blair et al.	Jan. 24, 1950
2,506,419	Graves	May 2, 1950
2,521,656	Segre et al.	Sept. 5, 1950
2,538,632	Tompkins	Jan. 16, 1951
2,547,874	Klema	Apr. 3, 1951

FOREIGN PATENTS

Number	Country	Date
567,280	Great Britain	Feb. 6, 1945