

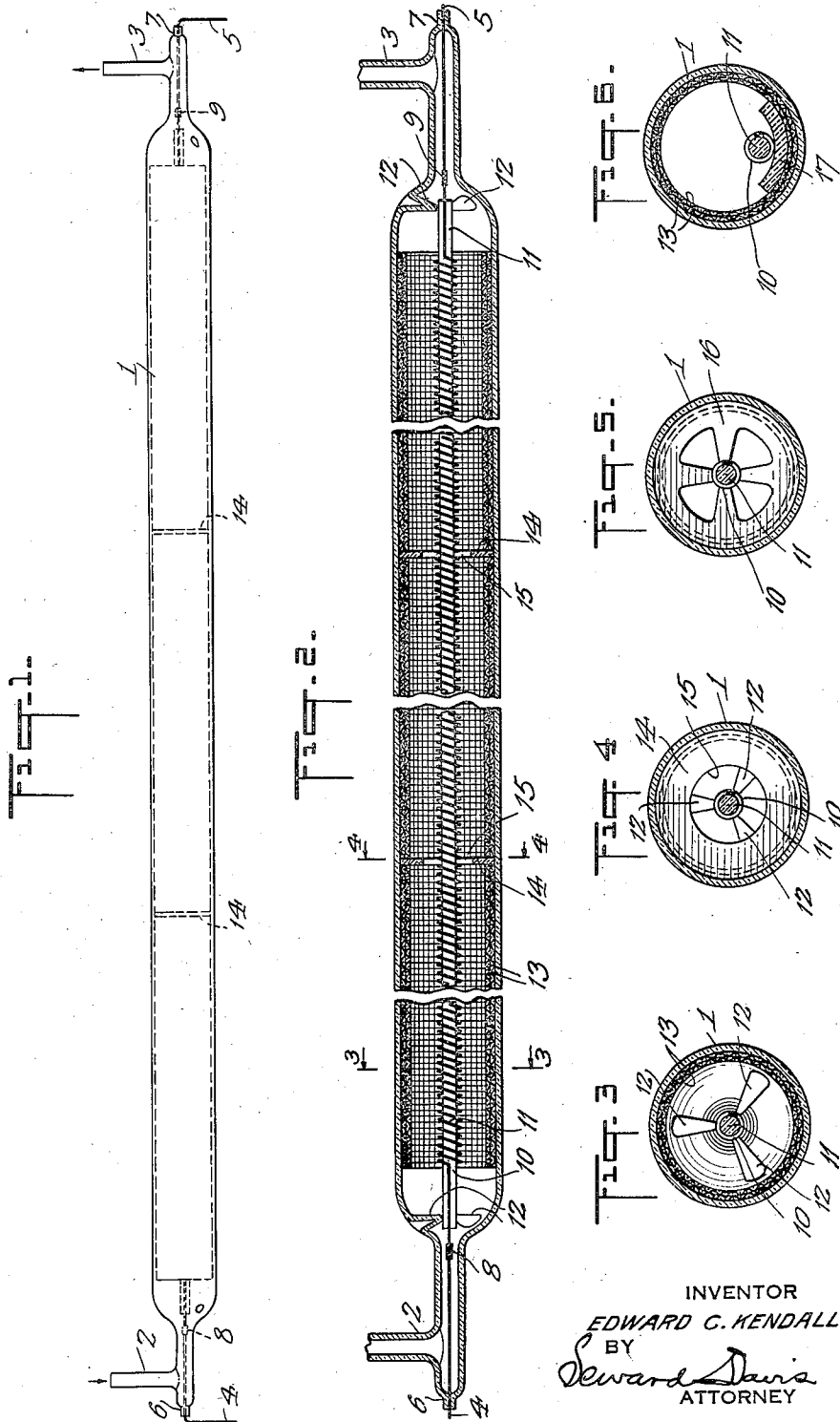
Aug. 21, 1934.

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1,970,700

APPARATUS FOR THE PURIFICATION OF GASES

Filed Dec. 13, 1930



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1,970,700

APPARATUS FOR THE PURIFICATION OF GASES

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Application December 13, 1930, Serial No. 502,122

5 Claims. (Cl. 23—234)

My invention relates generally to improvements in process of, and in apparatus for, the purification of gases containing traces of other gases capable of combining directly with certain metals at elevated temperatures; and more particularly to the purification of nitrogen from contained traces of oxygen or of oxygen and hydrogen.

The objects of my invention are: first, to provide a process for the treatment of gases for the complete removal of gaseous impurities therefrom; and particularly for such treatment of nitrogen containing traces of oxygen or of oxygen and hydrogen; second, the purification of one element of such a gaseous mixture as a continuous process; third, the practice of such process upon an industrial scale; and fourth, the provision of a simplified apparatus for the practice of such process.

The objects of my improvement in apparatus are: to provide a compact, self-contained and efficient furnace affording a free and continuous passage of the gas to be purified; to provide, in such a furnace, a reaction surface of maximum area of a metal adapted to combine or unite, at a certain elevated temperature, with the gas to be eliminated by such reaction; to provide a means of simultaneously heating by direct radiation the metal and the gas to the maximal temperature adapted to insure most rapid reaction but not sufficiently high either to decompose the resultant combination product or to impair the utility of the furnace elements; and to provide a furnace of the character described, in which all interior parts are visible during operation, thus permitting constant observation and checking up.

The foregoing and other and further features of advantage and utility hereinafter more particularly pointed out, are attained by the mechanism illustrated in the accompanying drawing forming a part of this specification, in which Figure 1 is an elevational view of the entire apparatus; Figure 2 is a view in vertical cross-section upon an enlarged scale; Figure 3 is a cross-sectional view on the line 3—3 of Figure 1, viewed in the direction indicated by the arrow; Figure 4 is a like view on the line 4—4 of Figure 1; Figures 5 and 6 are views similar to Figure 4 of modified forms of insulators adapted to support the heating element.

Similar numerals refer to similar parts throughout the several views, in which 1 indicates a tube of vitreous material such as pyrex glass; 2 is the gas inlet thereto and 3 the outlet therefrom; 4 and 5 are circuit connections to a source of suitable electrical energy. These con-

nections are sealed at 6 and 7, respectively, into the tube terminals; and at 8 and 9, respectively, are connected as by silver soldering, to an electrical resistance, such as the filament 10, preferably a coil of nichrome wire, wound upon the insulating support 11, which is preferably a rod of silica. The insulating support 11 may be in turn supported at its ends by the glass points 12, 12, preferably three at each end, formed by in-pressed portions of the tube 1. 13, 13, 13 are layers of metallic gauze or foil, preferably in cylindrical form, nested in the tube 1 and in contact with its wall. 14, 14 are insulating disks interposed between the adjacent ends of the rolls into which the layers 13, 13 are preferably formed, through which rolls and the openings 15 in said disks the resistance coil extends. Fig. 5 shows a disk 16 in spider form having the radial legs adapted to contact with the resistance coil and/or its supporting rod 11 and additionally support same in case of transportation or when the furnace is designed for use in other than its preferred horizontal position. Figure 6 shows a form of insulation alternative to the disk form, 17 being a strip extending lengthwise of the tube below the resistance coil.

Assuming, for example, that traces of oxygen are to be removed from nitrogen, or from carbon dioxide, then the layers 13 which present the reagent substantially in the all-surface condition of gauze or foil, will consist of a metal which is capable of combining directly with oxygen, such as copper, with which the oxygen will readily unite to form copper oxide.

Assuming, by way of further example, that traces of nitrogen are to be removed from carbon dioxide, or from argon, then a surface of a metal capable of uniting directly with nitrogen, such as magnesium, is provided, likewise in gauze or similar form, whereby magnesium nitride is formed.

Assuming, as a final example, that traces of oxygen and hydrogen are to be removed by a single treatment from nitrogen, the reaction element adapted for removal of oxygen, to-wit, copper, will have the functional efficiency to purify the nitrogen by removal of both oxygen and hydrogen.

With the proportionality of parts indicated in the drawing, and as hereinafter more explicitly detailed, the treatment of 500 c. c. of nitrogen per minute is contemplated, but it will be obvious to those skilled in the art that when a larger output is desired, the several structural elements are to be proportioned accordingly.

In prior devices of this art, a glass, crystal or similar tube, filled with copper, usually in wire form, has been used for the freeing of nitrogen from traces of foreign gases mixed therewith, by oxidation of such gases; but in such prior furnaces, the heat needed to initiate and promote the reaction has been applied to the exterior of the tube. These furnaces failed to attain the advantages inherent in the practice of the process involved in the use of my invention, for the reasons that the bulk of metal in the tube obstructed the free flow of gas therethrough. A further reason for the comparative inoperativeness of prior furnaces of the aforesaid type existed in the fact that the exterior heat was applied directly to the tube. As the desired temperature for reaction ranges from 400° to 600° or above, the external temperature impressed directly on the tube was so high as to soften it, resulting either in ballooning of the tube under the pressure of the gas or its collapse upon withdrawal of the internal pressure.

In practice with tubes of the prior type described, by reason of the ratio of mass of copper to its effective superficial area, the copper oxides resultant from the reaction of the oxygen and copper wire, were liable to be decomposed, as these furnaces were necessarily forced to run close to the maximum critical temperature to attain commercial efficiency.

To obviate these defects in theory and practice of the prior art, and to secure an operative furnace capable of attaining the hereinbefore recited objects, my invention consists in the provision of a heating element inside the tube, preferably parallel to the line of its axis; the provision of a reaction element consisting of the appropriate combining metal in a form best adapted to expose a maximum superficial area per unit of mass of such metal to contact with the heated gas stream; and so to relate such heating element and such reaction element that the latter will receive the direct radiation of heat from the former; and further so to locate the reaction element that the danger of excessive heating thereof may be avoided, while affording it the fullest opportunity for useful conversion of its absorbed heat in reaction with the gas to be eliminated.

The location of the heating element within the tube and in direct contact with the stream of gas freely flowing therethrough results in two distinctive features of operation of significant advantage as compared with prior structures: First, the degree of activation of the gas: The placement of the resistance element within the tube, with an unobstructed passageway afforded between it and the tube lining for the free flow of gas, renders it possible to raise the temperature of the gas higher than that of the copper lining. The reactivity of the gas surrounding the resistance element may be thus increased to a degree which can not be approached by the degree of activation of the gas obtainable with metallic copper within a tube which is externally heated, as in the old style furnaces. In such externally heated furnace the temperature of the gas in the tube can not be higher than that of the reactive material. The activation of the gas is not merely a matter of elevated temperature. Its degree of activation is a matter of temperature difference between the gas and the reagent, both being at elevated temperatures. Thus the activity of the gas increases as its temperature exceeds that of the reagent. By my

invention, the gas can be and normally is raised to a higher temperature than that of the surrounding coil of copper gauze which lines the tube. This higher temperature throws the gas into a violently agitated condition and forces it to react with the surface of the copper gauze under optimum conditions.

A further factor contributing to the attainment of this desired temperature differential is the area of contact of the metallic reagent with the tube. As a lining, in extended surface contact with the air-cooled tube, the copper gauze can be maintained below the temperature at which the oxygen is released from the copper oxide, while at the same time the gaseous mixture can be maintained at the desired maximum activation point. On the contrary, if the gas is heated by the metallic copper, as in the old type of furnace, then the product of the reaction, e. g. copper oxide, may be broken down by the heat into copper and oxygen. Therefore it is impossible to heat the gas adequately for maximum reaction and have the reaction product held in a stable form, if the heat is applied to the gas by means of metallic copper alone, as in prior furnaces. Second: the stability of the metallic oxide formed: The problem presented has two phases: (1) to heat the gas to a temperature necessary to make it combine with copper; (2) to supply a surface of metallic copper under conditions which will insure the formation of a stable compound, e. g. copper oxide, and bring about the removal of all traces of oxygen from the gas. In the old style furnace, attainment of these objects was attempted through the supply of heat to the copper within the tube from an external source. The result attained was a variable, whose efficiency depended upon the temperature, the velocity of the gas through the tube, and the condition of the copper within the tube. If this were heated to too high a temperature, it would fuse and become a solid block, whereby its entire surface would be tremendously reduced. If the tube was not heated sufficiently, then the gas was inadequately activated to combine with the copper. If it was heated to too high a temperature, then the copper oxide which had formed would subsequently break down with the liberation of oxygen. The arrangement of the tube according to my invention brings about a self-regulating distribution of temperature. The gas is heated much hotter than can be brought about in the old style furnaces, but the copper gauze which is placed in contact with the glass tube is maintained at a comparatively cool temperature. This insures the stability of the copper oxide which is formed, a feature which is not only novel but also of as significant advantage as the superactivation of the gases of the mixture to be purified.

The application of my invention to the attainment of these ends, in the case of the purification of nitrogen by removal of traces of oxygen present therewith, is contemplated in the structural embodiment illustrated in the drawing. This furnace consists of a pyrex glass tube, 34 mm. in outside diameter and 60 cm. in length, which is lined with a double roll of copper wire gauze of 40 mesh. The copper gauze roll fits snugly against the pyrex tube and is preferably divided into a plurality of sections. For the dimensions given, three units are employed, insulated from each other at their adjacent ends by mica disks whose outer peripheries contact with the inner wall of the pyrex tube. These disks are centrally

apertured to receive a resistance element comprising an insulating rod of silica upon which is wound a coil of nichrome filament 0.5 cm. in diameter, composed of 7.5 meters of number 18 nichrome wire. Leads, connected to the ends of the filament, extend through and are sealed in the tube terminals, adjacent which, respectively, are the inlet and outlet pipes for circulation of the gaseous mixture through the tube.

The operation of my furnace, as illustrated, is as follows:

The resistance element is heated by the passage of a suitably controlled current at a voltage adapted to raise the temperature of the air in the tube to approximately 400° C. Prior to its initial use for purification, it is desirable to pass a rapid current of air through the tube while heated to burn off oil and other alien substances coating the surface of the copper and to oxidize the copper, and then to pass a stream of hydrogen through the tube to decompose the copper oxide, the resultant water vapor being condensed and withdrawn. This pre-treatment results in an enormous increase in the surface of the copper because of the cracked and seamed surface resultant from the formation of the copper oxide, whose reduction with hydrogen does not greatly change the physical appearance or condition of this surface. This treatment may be repeated at occasional intervals with advantage to the maintenance of this surface in sensitive and highly reactive condition. The nitrogen purification may then be initiated by passage of gas therethrough at a volume flow of 200 c. c. per minute, and as the temperature is raised until it exceeds 400° C., the flow can be increased to 500 c. c. per minute. The nichrome coil may be raised to any temperature desired short of its decomposition; from 400° to 500° C. suffices to insure a high degree of reactivity of the traces of oxygen so that the oxygen rapidly combines either with the nichrome or the copper gauze; the temperature of the copper gauze is so high as to insure its rapid reaction with all traces of oxygen, but is not sufficiently high to drive off the oxygen from the copper oxide which has been formed. The contact of the copper gauze with the pyrex glass tube insures the maximal conduction of heat from the copper to the tube from which it is dissipated by the atmosphere, so that the resultant temperature to which the glass is heated does not exceed 300° C., which is too low to soften the glass, consequently the tube is not subject to severe thermal changes and will last indefinitely.

The furnace constructed as described is open for the free passage of gas, as the heating element and the copper gauze occupy a relatively small part of the cross section of the tube, and the velocity of the flow of the nitrogen stream through the tube is, therefore, not decreased. Thus a substantially constant flow, at the predetermined rate, of nitrogen heated to the maximal temperature affords a constant and uniform output of purified gas several times that obtainable either by (1) passing nitrogen through a solution which will remove the oxygen, such as alkaline pyrogallol, ammonium cuprous chloride and alkaline hydrosulfite, with or without a catalyst, as anthraquinone sulfonate; or by (2) removing the oxygen with a solution such as ammonium cuprous chloride placed in a large cylinder containing nitrogen under pressure; or by (3) passing nitrogen over a hot metal contained in a cylinder or tube heated externally, these

being the other methods and means heretofore practiced, so far as known to me.

Not only is the removal of traces of oxygen from nitrogen in the furnace herein set forth and described complete, but also it is possible to pass water vapor therethrough without the formation of even traces of hydrogen. Even if the water vapor is decomposed on the surface of the nichrome wire, the presence of the copper oxide completely removes all traces of the hydrogen so formed. In fact, all traces of hydrogen originally present in the nitrogen are removed as completely as is the oxygen.

The capacity of the apparatus constructed with the proportionalities specified is not strained by the rate of treatment involved in purifying 500 c. c. of gas per minute, since even after 24 hours of continuous operation, the copper oxide does not form for a distance of more than 4 cm. from the inlet end of the tube and not more than 15 cm. of the gauze is tarnished with the oxide.

Continuous use of the furnace brings about changes in the properties of the copper. It loses ductility and becomes highly reactive to oxygen. Its sensitivity is so increased that it will tarnish upon contact with atmospheric air at room temperature.

If the resistance coil is of a metal more readily susceptible to deformation under heat than nichrome, it may be found advantageous to support it, as is common practice in the use of copper as a resistance, by embedding it in vitreous material; but I find that normally the use of a filament of nichrome minimizes any necessity for other support than the silica rod for the resistance wire, and consequently for insulation to avoid a short circuit, as the tendency to deformation of the wire coil under heat is negligible at the temperatures involved. If the tube is operated as intended, i. e. in the horizontal position, the mica strip 17 extending lengthwise of the tube beneath the resistance, as shown in Fig. 6, may be substituted advantageously in lieu of the glass points 12 and/or spider 16.

Other structural modifications will suggest themselves as matters of proper engineering when operation of my invention upon a larger scale is undertaken, for example: by reason of the rapid oxidation of copper, the leads connecting the resistance may be of non-oxidizable metal or insulated from contact with the gas stream.

Having thus described my invention, I claim:

1. An apparatus for the removal of traces of an alien gas from a gaseous mixture, comprising a closed cylinder of vitreous material, an inlet thereto and an outlet therefrom; an electric circuit extending through said cylinder and including an electrical resistance axially disposed therein; a rigid support for said resistance; and a metallic lining contacting with said cylinder, the metal of said lining being capable of combining in the presence of heat with the gas to be removed to form a solid reaction product; the said cylinder affording an unobstructed passageway between said resistance and said lining from end to end.

2. An apparatus for purifying nitrogen including a tube of vitreous material, an inlet thereto and an outlet therefrom; terminals of an electric circuit sealed into said tube; an electrical resistance connected to said terminals, said resistance comprising a wire mounted upon a rod of insulating material; and a cylindrical roll of metallic gauze surrounding and spaced apart from said resistance and enclosed by said tube; where-

by a stream of gas may be continuously passed through said tube in contact with said resistance and be heated thereby, and whereby said roll may be heated by radiation from said resistance and by passage of the gas through the interstices of the roll, and whereby traces of oxygen mixed with said nitrogen may be caused to react with the metal of the roll to form oxides thereof.

3. An apparatus for purifying nitrogen including a tube of vitreous material, an inlet thereto and an outlet therefrom; terminals of an electric circuit sealed into said tube; an electrical resistance connected between said terminals, said resistance comprising a wire wound upon a rod of insulating material; a plurality of rolls of copper gauze at spaced intervals along and surrounding but spaced from said resistance and enclosed in and supported by said tube; and insulating disks at spaced intervals between the adjacent rolls of gauze; whereby a stream of gas may be continuously passed through said tube in contact with said resistance and be heated thereby, and whereby said rolls may be heated by radiation from said resistance and by passage of the gas through the interstices of the rolls, and whereby traces of oxygen mixed with said gas may be caused to react with the copper of the rolls to form oxides thereof.

4. An apparatus for purifying nitrogen including a tube of glass, an inlet thereto and an outlet therefrom; terminals of an electric circuit sealed into said tube; an electrical resistance connected to said terminals, said resistance including a fila-

ment of resistance wire mounted upon a rod of insulating material; a cylindrical roll of metallic gauze surrounding but spaced from said resistance and enclosed by said tube; and insulating supports for said rod and/or said filament; whereby a stream of gas may be continuously passed through said tube in contact with said resistance and be heated thereby, and whereby said roll may be heated by radiation from said resistance and by passage of the gas through the interstices of the roll, and whereby traces of oxygen mixed with said gas may be caused to react with the metal of the roll to form oxides thereof.

5. An apparatus for purifying nitrogen including a tube of vitreous material, an inlet thereto and an outlet therefrom; an electric circuit extending through said tube and including an electrical resistance, said resistance comprising a filament mounted upon a rod of silica; a cylindrical roll of copper gauze surrounding and spaced from said resistance and enclosed and supported by said tube; and insulating supports for said rod and said filament; whereby a stream of gas may be continuously passed through said tube in contact with said roll and be reacted upon thereby, and whereby said roll may be heated by radiation from said resistance and by passage of the gas through the interstices of the roll, and whereby traces of oxygen mixed with said gas may be oxidized by reaction.

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