B. E. ELDRED.

CATALYTIC BODY.

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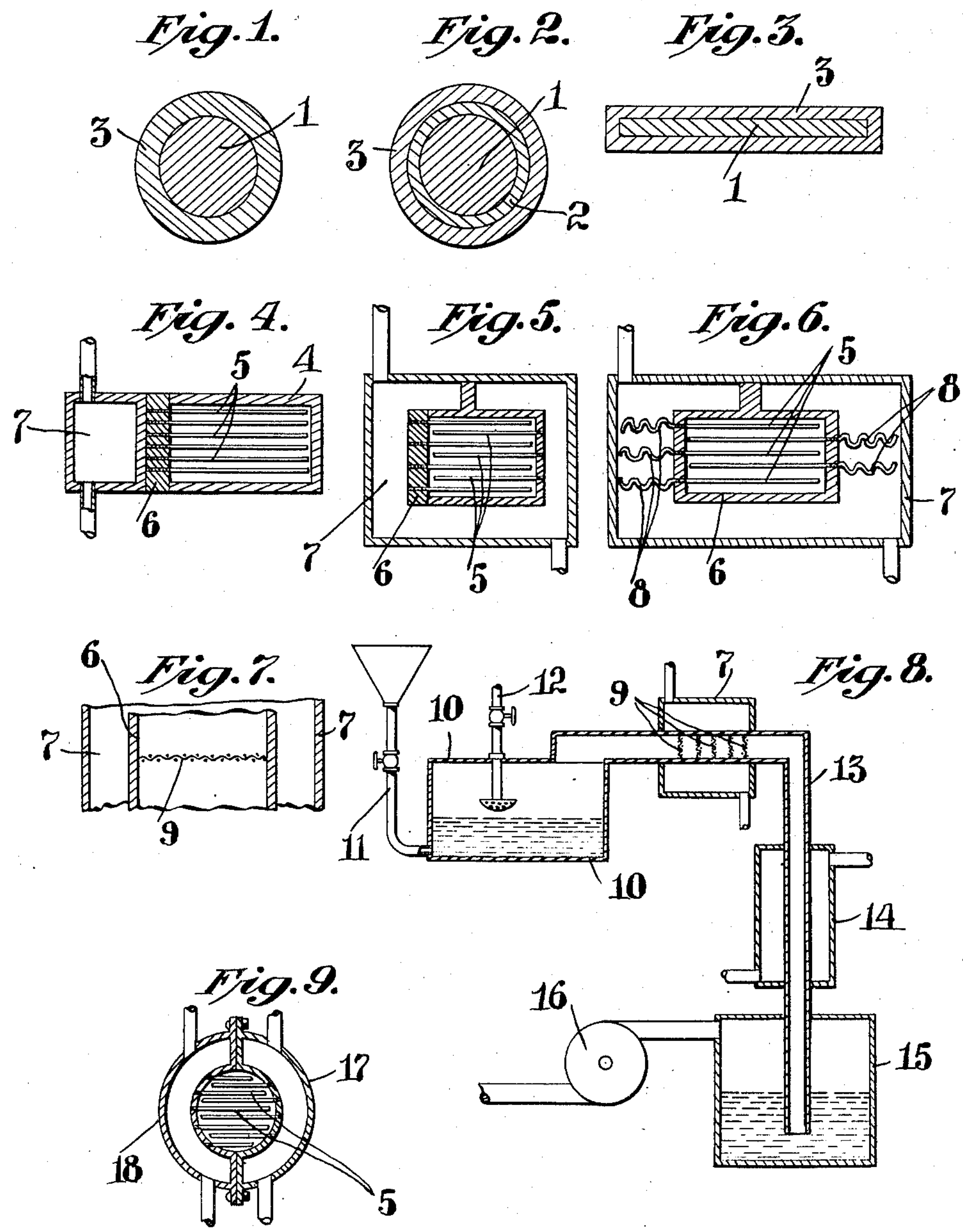
1,043,580. Patented Nov. 5, 1912.



Attest:

by

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UNITED STATES PATENT OFFICE.

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CATALYTIC BODY.

1,043,580. Specification of Letters Patent. **Patented Nov. 5, 1912.**

Application filed January 7, 1908, Serial No. 409,622. Renewed May 9, 1912. Serial No. 696,279.

To all whom it may concern:

Be it known that I, BYRON E. ELDRED, a citizen of the United States, residing at Bronxville, in the county of Westchester and State of New York, have invented certain new and useful Improvements in Catalytic Bodies; and I do hereby declare the following to be a full, clear, and exact description of the same, such as will enable others skilled in the art to which it appertains to make and use the same.

This invention relates to catalytic bodies and consists in catalytic bodies or devices comprising compound metal bodies having a core of one metal and an enveloping sheath of another and catalytically acting metal, such core and sheath being firmly and permanently welded or united together with a cohering union in such manner as to assure both mechanical strength to such union and good thermal conductivity between said joined metals; all as more fully hereinafter set forth and as claimed.

Many metals possess what is called a catalytic power of promoting reactions in mixtures of gases or liquids contacted with them, as in the action of platinum in bringing about limited or complete oxidation of mixtures of combustible gases or vapors with oxygen, the hydrogenating action of nickel and cobalt in causing the union of hydrogen with various carbon compounds, etc. Since the amount of such catalytic action performed in a given time unit in a body of gas is strictly proportionate to the exposed surface of catalyzing metal, it is customary to use such metals in finely divided form, sometimes as masses of powder and sometimes as powders adhering to and held by inert porous materials, such as asbestos, glass wool and the like. These expedients however while giving great surface to a given amount of metal, do not give a proportionately great exposure of such metal to the gases or vapors to be treated. It is substantially impossible to drive or distribute gases uniformly throughout a body of powder, and in passing gases over a body of such powder it is substantially only the top layers of the powder which display a maximum activity, underlying layers not functioning to any great extent. Use of very thin layers of powder of course necessitates unduly extended shelf surface. Incorporation of the finely divided metal in asbestos or glass wool works better, but since such powders do not as a rule adhere well to the carrier it is necessary to employ such carrier rather closely packed or in a form which renders difficult the circulation of gases therethrough. Impregnation of the carrier with the metal and formation of the carrier into balls or pellets is therefore resorted to, but in this manner of operation the metal in the interior of a ball or pellet is of relatively little use. Another and more serious difficulty with the described ways of using catalytic metals consists in the difficulty of temperature regulation. Nearly all catalyses are exothermic reactions, heat being developed by the action, and frequently the amount of heat is rather large. And as it is usually desirable to work within comparatively narrow temperature limits, keeping and maintaining the catalytic metal within a few degrees of some definite temperature, this evolution of heat is a serious matter. Nearly all the catalytically acting metals are relatively poor conductors of heat, even in the reguline form, and in the form of powders they are worse. It is difficult to keep a body of platinum powder at any definite temperature, and when the platinum is distributed through a mass of such a heat-insulator as asbestos, it is very hard to prevent the accumulation of reaction heat in the metal.

It is the purpose of the present invention to obviate the described difficulties by the provision of catalytic bodies comprising the catalytic metal in the form of very thin continuous layers or films carried by bodies of better heat-conducting metals weld-united to such layers or films and therefore in absolute metallic union therewith whereby by controlling the temperature of the carrying metal the temperature of the film or layer can also be controlled. Being so thin, the heat conducting qualities of the film metal are more or less immaterial. It is obvious that with such very thin films, a given weight of the catalytic metal can be given an enormous surface while by carrying the film on a substantial body of the core metal the compound catalyst can be given shapes or forms suited to induce thorough contact with a current of gas or vapor, as in using the catalyst in the form of wire gauze septa or diaphragms across gas-carrying conduits, by the use of shaped gauze or leaf-metal wings or vanes in such conduits in forms adapted to produce vortices in passing gases, or by the use of staggered sheet metal partitions in such conduits. In whatever form the compound metal catalyst be employed, it is desirable that it be in thermal contact with heat-interchanging elements in or on the vessel carrying or containing the gases or vapors to be catalyzed so that definite temperatures may be maintained in such catalyst. If the catalyst be in the form of wires, for instance, such wires may be threaded at their base into a larger body of metal or they may be butt-welded to copper and similar wires or rods extending into another chamber carrying a heating or cooling fluid.

Suitable catalysts under the present invention may be made by welding a sheath or coating of platinum on a billet of copper or steel or soft iron, and coextending the joined metals to form wire or sheet metal. If 3 to 10 per cent. of platinum be placed on the original billet and coextension be performed with care, the wire, sheet or leaf metal formed will also have 3 to 10 per cent. of platinum but this thickness in such coextended ware will correspond to an extremely tenuous layer. Steel and platinum do not readily unite directly, but as I have discovered, their union can be produced by contacting clean platinum with clean molten steel; one method of doing this being set forth in my companion application Ser. No. 409,617, filed Jan. 7, 1908. Unions produced in this manner are, or are equivalent to, weld unions, and the joined metals can be coextended in the same manner as steel alone. By using a soft steel or pure iron, such as, for instance, that produced in the Tropenas process, very good thermal conductivity can be obtained in the core metal. A pure iron core being shielded by the platinum sheath, the joined metals can be worked together without contamination of such iron; a matter which is, with naked pure iron, rather difficult. Copper cored platinum can be produced by contacting hot clean copper, either molten or near the melting point, with clean platinum as disclosed in my copending applications Ser. No. 409,618 and Ser. No. 409,620, both filed Jan. 7, 1908. Gold and silver cored platinum may be produced in similar manner. As copper, gold and silver unite with platinum with less difficulty than does steel, the steel may be preliminarily filmed or coated with one of these metals prior to applying the platinum. Steel permanently, firmly and autogenously united to coatings of copper, silver or gold may be produced by the method described by J. F. Monnot in his Patent No. 853,716, and such coated steel may be employed in producing platinum coated billets which may then be drawn, rolled or otherwise extended to form the platinum filmed catalysts of the present invention.

Cobalt and nickel may be united to steel or copper billets, and the duplex or compound billets extended in similar manner to produce catalysts having film coatings of cobalt or nickel. The cobalt or nickel may be united to the underlying core metal directly or through a linking layer of another metal such as gold, silver or copper. The use of such a linking layer, apart from its convenience in many cases in producing the desired type of union, has further substantial advantages for the present purposes. In use, the catalytic metals are apt to become more or less porous or fissured, and, with the film coatings of the present invention, avenues may be opened whereby corrosive gases or liquids may gain access to the underlying core metal. In some catalyses where corrosive bodies are produced, as in the union of sulfur dioxid with oxygen to produce sulfur trioxid, if the core metal be readily corrodible, as is the case with steel, this phenomenon may be disadvantageous. Gold and silver not being catalytic metals to any extent are not subject to this fissuring or porosity, and by using either as a linking layer, the core metal may be kept hermetically sealed. The same is true of copper, though to a rather less extent. It is however very well adapted for cases where the corrosive body produced is, as in the case of oxidizing alcohol to acetic acid, only feebly corrosive.

The core metal may be, and very advantageously, either gold or silver. Both are highly conductive for heat. Using either metal as a linking layer between platinum and iron, they serve to convey and equalize the heat in the core. The same is true of copper as a linking layer.

The porosity of the catalytic film may be purposely produced, and particularly where a linking layer is employed with a ferrous metal core, or where a copper, gold or silver core is employed. In the case of platinum, this may be done by treating the platinum-filmed catalysts with corroding agents, either liquid or gaseous, such as *aqua regia* or bromin water or chlorin gas, and subsequently heating. The attack of these reagents upon the film should of course not be prolonged. In the case of catalysts carrying filmed cobalt or nickel, the catalysts may be slightly oxidized by heating in air and then reduced in a current of hydrogen or other combustible gas.

Catalysts according to the present invention may be employed in a wide variety of catalytic reactions, such as the production of aldehydes and acids by limited oxidation of alcohols in the presence of air, as in the formation of acetaldehyde or acetic acid from ethyl alcohol or formaldehyde from methyl alcohol, the production of sulfur trioxid by the oxidation of sulfur dioxid, ignition of combustible gases, as in self-lighting gas burners, the hydrogenation of carbon compounds by cobalt and nickel, as in reducing carbon monoxid or dioxid to methane in the presence of hydrogen, the formation of isopropyl alcohol by catalyzing a mixture of hydrogen and acetone, the formation of hexhydrobenzol from benzol and hydrogen, etc. In these reactions, as stated, heat is generally produced while it is desirable that the catalyst be maintained at a tolerably uniform temperature. It is for this reason that the catalyst should generally be maintained in thermal contact with a mass of metal provided with heating or cooling means.

In the accompanying more or less diagrammatic illustrations, I show sundry embodiments of the described invention.

In this showing, Figure 1 illustrates a cross-section (dimensions being exaggerated) of a wire composed of a core 1 of iron, copper, gold, silver, etc., having united thereto a film 3 of a catalytic metal such as platinum, palladium, nickel, cobalt, etc. Fig. 2 illustrates a similar view of a wire having core and catalytic film sheath linked by an intermediate layer, 2, of a conductive, non-catalyzing metal such as gold, silver or copper, weld-united on either side to core and sheath. Fig. 3 is a similar view of a sheet or leaf of compound metal composed of core 1 and sheath 3. Fig. 4 is a cross-section of an apparatus utilizing the catalysts of the present invention, 4 being a conduit for carrying gases or vapors to be catalyzed, 5 compound metal wires spanning the same, 6 a body of metal to which they are united and 7 a heat-interchanging jacket; Fig. 5 is a similar view in which the heat interchanger surrounds the gas conduit and the wires are staggered; Fig. 6 is a similar view in which the catalyzing wires in lieu of being secured at their base, as by soldering or butt-welding, to the heat interchanger wall, are butt-welded to copper wires 8 extending out through such wall into the heat interchanger chamber; Fig. 7 is a cross-section of a fragment of apparatus in which the catalyzing wires are formed into a gauze septum or diaphragm 9 spanning the gas conduit 6 and soldered or otherwise secured to its walls. Fig. 8 is a view in vertical cross-section and partial elevation of a complete apparatus employing the described catalyzers, a liquid vessel 10, provided with valved liquid inlet 11 and valved gas inlet 12, communicating with a catalyzing conduit having catalytic gauze septa spanning it and in thermal relationship to a heat-interchanging jacket. Beyond the catalyzing conduit is a pipe 13, passing through a condenser jacket 14 into a seal-pot 15 from which uncondensed gases may be withdrawn by fan 16. Fig. 9 is a detail view showing an advantageous method of constructing the gas conduit, such conduit being formed of two half-tubes, 17 and 18, each carrying catalyzing wires and each provided with a thermal jacket.

In the use of the structure of Fig. 8, the liquid in 10 may be methyl alcohol and a regulated amount of air sucked in through the gas inlet by the fan shown. This air coming into contact with the alcohol evaporates part of it and the mixture of air and vapor passes into intimate contact with the filmed catalyst on the wire-gauze septa shown, producing methyl aldehyde. Circulation of a suitable thermal fluid in the jacket serves to control the temperature of the filmed catalytic metal by thermal conduction through the core metal. For producing formaldehyde in this manner, the catalytic metal is preferably platinum and the core metal copper or steel. The formaldehyde passes through the condenser and accumulates in the body of liquid in the seal pot. For making isopropyl alcohol, the liquid may be acetone and the gas introduced hydrogen, while the catalytic metal is preferably nickel or cobalt. The thermal jacket should be used in this case to maintain a temperature quite a little above the boiling point of water. With a nickel catalyst, the liquid may be benzol (C_6H_6) and the gas hydrogen, hexahydrobenzol being produced.

In producing sulfur trioxid, the catalytic body may be very advantageously filmed platinum carried by a copper core with a linking layer of gold or silver between, rapid exchange of heat being very necessary in this instance. In forming methane from water gas, etc., the catalyst is advantageously nickel filmed on steel or soft iron.

In producing the catalytic bodies, it is inadvisable to unite the coating of catalytic body to the original billet by any form of solder, solder not standing the great extension necessary to produce the desired filmiform catalytic metal. On the other hand, when the metals are actually welded together the coating catalyst may be extended to a tenuity at which it would be utterly unable to exist alone, as it can rely on the welded core for support. With a weld-union, thermal conduction is perfect.

Where, as frequently happens in catalyses, the amount of heat developed in the reaction is insufficient to maintain the apparatus at the desired temperature, heat may of course be supplied to the catalyst through the intermediancy of the heat interchanger.

What I claim is:—

1. In a catalyzing apparatus, the combination of a catalytic body comprising a core of relatively heat-conductive metal provided with a filmiform continuous coating layer of a relatively less heat-conductive catalytic

metal in heat-conducting relationship thereto and heat-controlling means in thermal relationship to said catalytic body.

2. In a catalyzing apparatus, the combination of a catalytic body comprising a core of a relatively heat-conductive metal provided with a filmiform continuous coating layer of a relatively less heat-conductive catalytic metal in heat-conducting relationship thereto, said coating layer and said core metal being metallically united to an intermediate linking layer of another metal, and heat-controlling means in thermal relationship to said catalytic body.

3. A catalytic apparatus provided with an element comprising a core of heat conductive metal carrying a continuous filmiform layer of platinum in heat conducting relationship thereto.

4. A catalytic body comprising a core of heat conductive metal carrying a continuous filmiform layer of platinum in heat-conducting relationship thereto, both said metals being weld-united to an intermediate linking layer of another metal.

5. In a catalyzing apparatus, the combination of a catalytic body comprising a core of a relatively heat-conductive metal provided with a continuous filmiform coating layer of a relatively less heat-conductive catalytic metal in heat-conducting relationship thereto, the surface of said catalytic metal being porous, and heat-controlling means in thermal relationship to said catalytic body.

6. A catalytic body comprising a core of heat conductive metal carrying a continuous filmiform layer of catalytic metal in heat conducting relationship thereto, both said metals being weld-united to an intermediate impervious linking layer of another metal and the surface of the catalytic metal being porous.

7. In a catalyzer, the combination of a catalytic body comprising a core of heat conductive metal carrying a filmiform layer of a relatively less conductive catalytic metal in heat conducting relationship thereto with a heat interchanger in heat-conducting relationship with said catalytic body.

8. In a catalyzer, the combination of a catalytic body comprising a core of heat conductive metal carrying a filmiform layer of a relatively less conductive catalytic metal in heat conducting relationship thereto and having an intermediate linking layer of another metal weld-united to both such metals, with a heat interchanger in heat-conducting relationship to said catalytic body.

9. In a catalyzer, the combination of a catalytic body comprising a core of heat conducting metal carrying a filmiform layer of platinum in heat conducting relationship thereto, with a heat interchanger in heat conducting relationship to said catalytic body.

10. In a catalyzer, the combination of a catalytic body comprising a core of heat conducting metal carrying a filmiform layer of platinum in heat conducting relationship thereto and having an intermediate linking layer of another metal weld united to both metals, with a heat interchanger in heat conducting relationship to said catalytic body.

In testimony whereof I affix my signature, in the presence of two witnesses.

BYRON E. ELDRED.

Witnesses:
K. P. McElroy,
H. M. Marble.