Sept. 9, 1958 C. J. COBERLY ET AL 2,851,340

APPARATUS FOR PRODUCING ACETYLENE BY THE PYROLYSIS OF A SUITABLE HYDROCARBON

Filed March 4, 1952

2 Sheets-Sheet 1

Fig. 1.

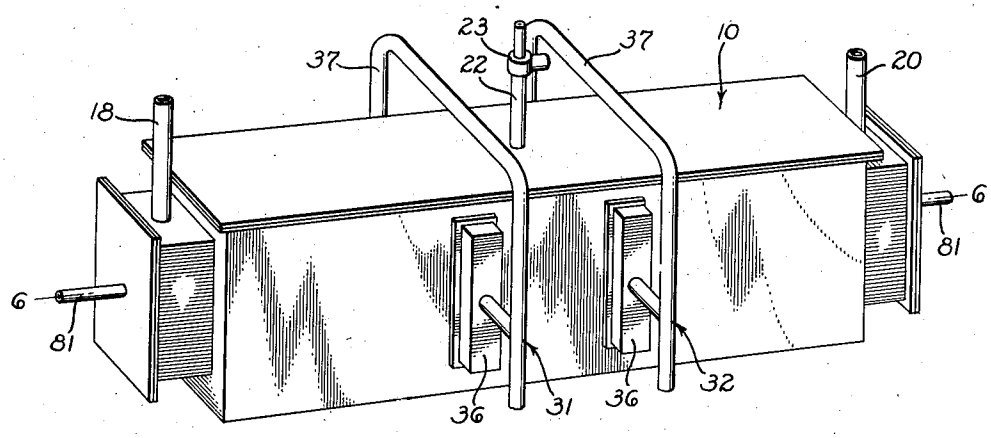


Fig. 2.

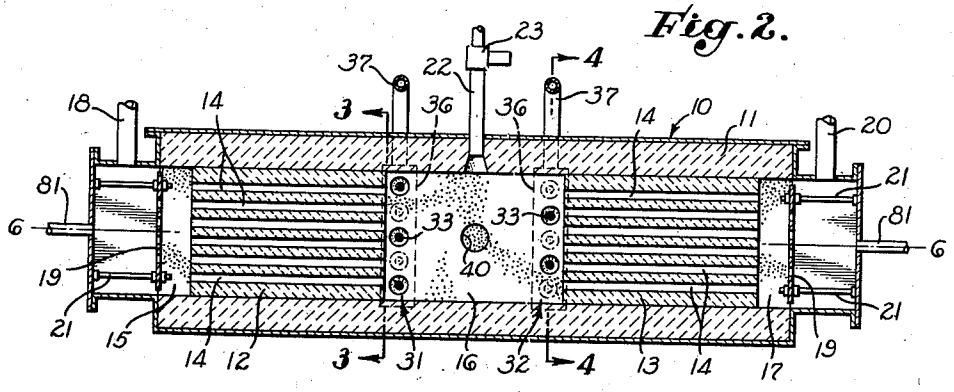
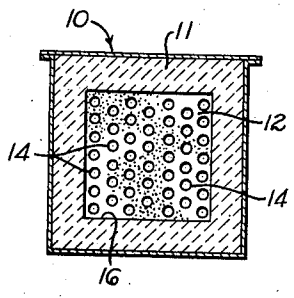


Fig. 3.



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Fig. 4.

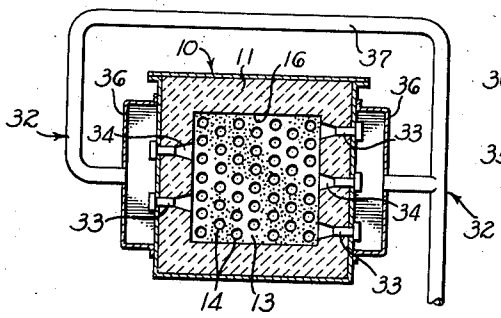


Fig. 5.

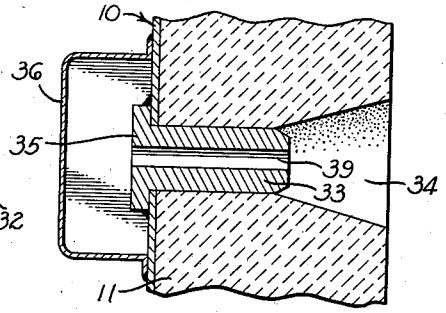
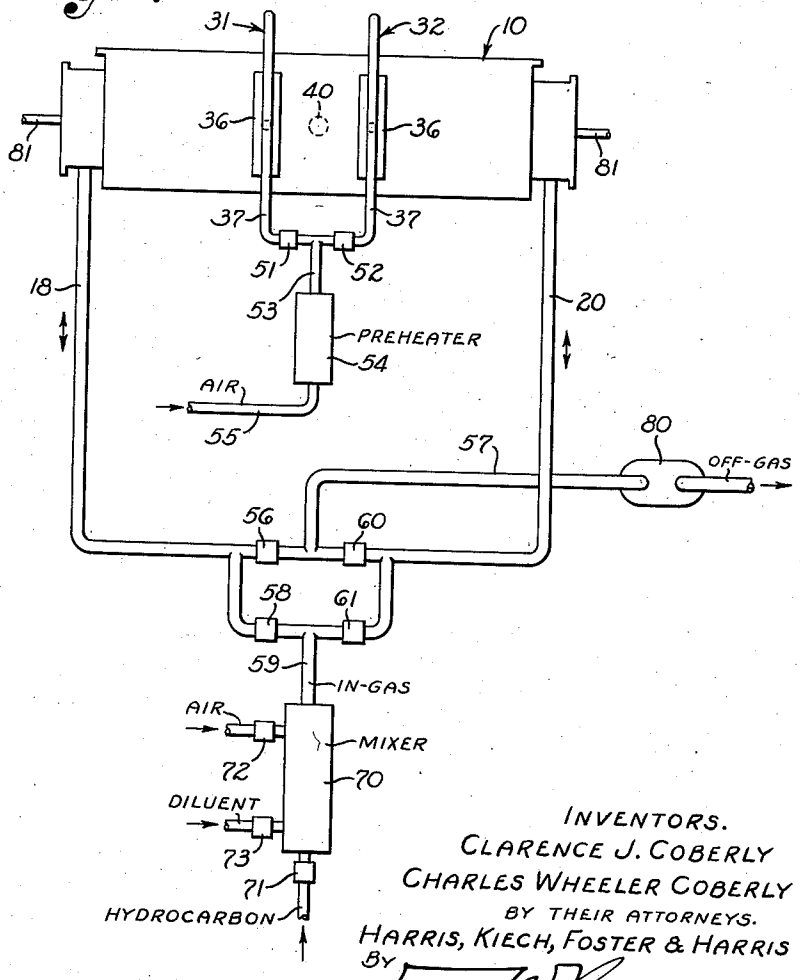


Fig. 6.



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APPARATUS FOR PRODUCING ACETYLENE BY THE PYROLYSIS OF A SUITABLE HYDROCARBON

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2 Claims. (Cl. 23—277)

Our invention relates to the art of producing acetylene by the pyrolysis of other hydrocarbons. It is well known in the art that if a suitable hydrocarbon is raised to reaction temperature at least a portion of the hydrocarbon will be converted to acetylene, but it is also well known that if the hot acetylene is then cooled slowly, the acetylene decomposes into free carbon and hydrogen, releasing considerable heat.

It is an object of our invention to provide a process by, and an apparatus in, which acetylene can be formed by the pyrolysis of a suitable hydrocarbon and by, and in, which the acetylene so formed can be recovered with very little, if any, decomposition.

To produce acetylene by the pyrolysis of a suitable hydrocarbon, large amounts of heat must be supplied to produce the necessary endothermic reaction, this heat being hereinafter referred to as reaction heat which, of course, is eventually latent heat in the acetylene. An almost equal amount of heat is needed to raise the in-gas to reaction temperature, this heat being hereinafter referred to as sensible heat. This heat is carried as sensible heat in the off-gas produced by the reaction and must be rapidly removed from the off-gas by cooling the off-gas, if any substantial proportion of acetylene is desired as a final product.

It is a further object of our invention to provide a process and an apparatus in which the off-gas is not only rapidly cooled, but in which a large proportion of the sensible heat so removed from the off-gas is retained in the apparatus and re-used to supply sensible heat to the in-gas.

It is not particularly difficult to provide a process and apparatus which will produce an off-gas containing some acetylene but it is only by providing an apparatus having definite proportions and characteristics and operating this apparatus within definite limits that an off-gas containing a substantial proportion of acetylene can be produced.

It is a further object of our invention to provide an apparatus of such proportions and characteristics, and to provide a process operating within such limits, that an off-gas containing a substantial proportion of acetylene can be produced from an in-gas containing a substantial proportion of a suitable hydrocarbon.

The term "suitable hydrocarbon," as used herein to denote a hydrocarbon capable of producing acetylene, is hereby defined as including all those hydrocarbons, and limited to only those hydrocarbons, which were known in the art, at the time the application for this patent was filed, to be capable of forming acetylene by pyrolysis. Any such hydrocarbon can, of course, be used, but the selection of this hydrocarbon depends upon availability and cost. Natural gas, at points where it is available, is at this time probably the cheapest suitable hydrocarbon, but butane, popane and ethane are more economical in some localities.

The term "substantial proportion," wherever used herein to denote the proportion of gas in any mixture,

is hereby defined as a proportion of at least two percent (2%) by volume of the total mixture.

The term "in-gas" is hereby defined as a gas consisting largely or partially of a suitable hydrocarbon which is delivered to the reaction zone in which conversion to acetylene occurs.

The term "off-gas" is defined as the gas mixture which contains acetylene as it leaves said reaction zone.

The term "ignition temperature," as used herein, is defined as the temperature at which the suitable hydrocarbon will break down to release hydrogen and thus initiate combustion in the presence of oxygen. Propane so breaks down at about 600° C. and methane breaks down at about 900° C. The ignition temperature of practically all suitable hydrocarbons is known in the art or can be easily determined by a person skilled in the art.

The term "reaction temperature," as used herein, is defined as the temperature at which the suitable hydrocarbon is molecularly converted from its original state into acetylene. The reaction temperature of propane is about 900° C. and the reaction temperature of methane is about 1200° C. The reaction temperatures of many suitable hydrocarbons are known in the art or can be easily determined by a person skilled in the art.

The term "non-flammable mixture" is used herein to denote a mixture of gases containing a hydrocarbon and oxygen in which the amount of oxygen present is substantially less than that needed for complete combustion of the hydrocarbon.

"Complete combustion" of a hydrocarbon, wherever used herein, is defined as a combustion in which the hydrocarbon is converted to carbon monoxide, carbon dioxide and water. "Incomplete combustion" of any in-gas which contains a hydrocarbon is defined as a combustion which leaves some hydrocarbon in the off-gas.

Further objects and advantages will be set forth hereinafter.

The apparatus used is disclosed in Figs. 1 to 6 in which:

Fig. 1 is a perspective view of this furnace;

Fig. 2 is a section therethrough;

Fig. 3 is a section therethrough on a plane represented by the line 3—3 of Fig. 2, this plane being viewed in the direction of the arrows adjacent the ends of this line;

Fig. 4 is a section therethrough on a plane represented by the line 4—4 of Fig. 2, this plane being viewed in the direction of the arrows adjacent the ends of this line;

Fig. 5 is a section through the wall of the furnace and through a gas nozzle projecting into a hole through said wall; and

Fig. 6 is a diagram of the apparatus including furnace, valves and piping.

In the following specification and claims, the abbreviations "RH" for right-hand and "LH" for left-hand are freely used to denote the position of parts as seen in the drawings.

The furnace shown in the drawings is primarily one in which high temperature endothermic reactions may be carried on. The furnace is well adapted to the conversion of any suitable hydrocarbon to another hydrocarbon, for example, the conversion of methane to acetylene.

The furnace consists of a gas-tight steel shell **10** having a heat refractory lining **11** of fire brick or the like. Placed inside are two refractory masses, an LH mass **12** and an RH mass **13**. The masses may be made of alundum silicon carbide or zirconium oxide. The masses must be made of material that, over long periods, will withstand temperatures somewhat in excess of 1500° C. without material injury. The masses **12** and **13** have small cylindrical channels **14**, shown in Fig. 3, extending therethrough in a direction substantially parallel to the major axis **6—6** of the furnace. These channels **14** may

be one-half inch (½ in.) or less in diameter and are substantially straight and unimpeded. The two masses **12** and **13** divide the interior of the furnace into three chambers, namely, an LH end space **15**, a central space **16**, and an RH end space **17**.

An LH off pipe **18** takes or delivers gas to the LH end space **15** and an RH off pipe **20** takes or delivers gas to the RH end space **17**. A perforated dispersion plate **19** spaced away from the inside end of the shell on rods **21** is provided at both ends of the furnace to give a more even distribution of the in-gas to the channels **14** in both masses **12** and **13**.

A central off pipe **22** having a valve **23** takes gas from or delivers gas to the top of the central space **16**. Projecting into each side of the furnace in a plane close to the inner face of each of the masses **12** and **13** is a series of nozzles. The LH series **31** is in a plane close to the inner or RH face of the LH mass **12**, and the RH series **32** is in a plane close to the inner or LH face of the mass **13**. In a small furnace there will be five nozzles **33** to each series of nozzles **31** and **32**, three on one side of the furnace and two on the other, as shown in Fig. 4 of the drawings. Each of the nozzles **33**, as shown in Fig. 5, is inserted in an opening **34** in the lining **11** of the furnace, the head **35** of the nozzle being welded to the shell **10** of the furnace inside a manifold **36** which is also welded to the shell **10**. The two manifolds of series **31** and **32**, one on each side of the furnace, are connected by pipes **37** so that all the nozzles **33** of each series may be supplied with gas through a suitable valve. Gas under pressure is jetted through the small orifices **39** of the nozzles **33** at high velocity and forms a sheet of gas parallel to the end of each mass and the gas from the channels **14** impinges on this sheet at right angles thereto. We have found that this arrangement greatly facilitates rapid and complete mixing of the gases which is necessary in any reaction which may be performed in the furnace. In making acetylene, the total time the mixed gases are at reaction temperature must be less than one-tenth (1/10) of a second. An opening **40** provided with a tight lid or cover is provided in the side of the furnace so that a torch may be used to initiate combustion in the central space **16** whenever the furnace is started cold.

We have found it necessary to place the furnace with its major axis horizontal if it is to be used with high temperature reactions taking place in the central space **16**. By placing the furnace with its axis horizontal and with the channels **14** also horizontal, the hot masses **12** and **13** may be supported on their sides on the lining **11** and no metal supports for the masses **12** and **13** are necessary. This is important since any metal of reasonable cost would be melted or perhaps vaporized at furnace temperatures, or might act catalytically to produce undesirable results.

The furnace shown and described herein may be used for many purposes, particularly, for example, to produce acetylene from a suitable hydrocarbon, for example, methane, ethane, propane, or butane. Acetylene can be so produced by various processes which may be conducted in this furnace, for example, by a process which may be described as follows.

When the furnace above described is to be used to conduct this particular process, additional parts must be provided to make a complete apparatus which may be described as follows.

The pipes **37** forming parts of the nozzle assemblies **31** and **32** are connected through valves **51** and **52** to an air pipe **53** which is preferably connected to a preheater **54** which is supplied with cold air through a pipe **55**. Preheated air is not absolutely essential to the process and the heater **54** will probably not be used in many installations. The pipe **18** may be connected through a valve **56** with a off-gas pipe **57** or through a valve **58** with an in-gas pipe **59**. The pipe **20** may be connected through a valve **60** with the off-gas pipe **57** or through a valve **61** with the in-gas pipe **59**. In-gas is produced in a mixer **70**, a suitable hydrocarbon being delivered to the mixer **70** through a valve **71**, air being delivered through a valve **72** and a diluent such, for example, as steam being delivered through a valve **73**.

Gases may be drawn from the apparatus through the pipe **57** by an exhauster **80** which may pull a substantial vacuum on the interior of the furnace shell **10**. In some furnaces it is desirable to inject water into the spaces **15** and **17** through pipes **81**.

Suitable thermocouples, not shown, are provided at various points in the furnace, and suitable timing devices are provided for actuating the various valves, all of which are power operated. The furnace is heated as a preliminary step to cyclic operation. This heating can be accomplished by starting the exhauster and opening at least one of the valves for example the valve **56**, all other valves being closed, and inserting a torch through the opening **40** so that the products of combustion from the torch are drawn from the central space **16** through the LH mass **12** and out through the pipe **18**, the valve **56** and the pipe **57** by the exhauster **80**. This heating is ordinarily conducted slowly which is not objectionable since once heated the furnace may be operated under cyclic conditions over a long period without reheating. In practice a more elaborate arrangement may be provided for preheating but since this preheating forms no part of the invention, it need not be more specifically described.

The cyclic operations may be described as starting with the LH mass **12** preheated so that its RH end is at a temperature above 1200° C. and its LH end is at a substantially lower temperature. The cycle will consist of an RH flow in which gas flows from the space **15** toward the space **17** and an LH flow in which gas flows from the space **17** toward the space **15**. During the RH flow the valves **51**, **58**, **60**, **71**, **72**, and **73** are open and the exhauster **80** is drawing gas through the furnace. The valves **71**, **72**, and **73** which are left open during the entire cycle are set to produce an in-gas in the pipe **59** consisting of the suitable hydrocarbon, sufficient air or oxygen to provide for the complete combustion of only a portion of the suitable hydrocarbon, plus some steam.

In large plants oxygen may be supplied through the valve **71** and through the pipe **55** instead of air, in which case the amount of oxygen supplied must be correspondingly restricted. The in-gas delivered through the pipe **59** is non-flammable in small passages such as the channel **14** when cold, but it will burn when the entire mass of the in-gas is heated above the ignition point. It is so heated by transfer from the LH mass and when it has been heated above the ignition temperature in channels **14**, a limited and partial combustion occurs, followed by a partial decomposition of the hydrocarbon. The amount of air or oxygen injected through the nozzles **33** may be sufficient to provide for the complete combustion of from 10% to 40% of the suitable hydrocarbon delivered to the mixer **70** through the pipe **71**.

The temperature in the central space **16** will then probably not exceed 1300° C. with propane in-gas, or 1500° C. with methane in-gas. At these temperatures a portion of the hydrocarbon is converted into acetylene. It is important, however, to operate the apparatus so that there is only a partial conversion and the off-gas contains a substantial proportion of hydrocarbons other than acetylene. In other words, the furnace must be operated so there is not only partial combustion of the hydrocarbon, but also this partial combustion must be controlled to give a maximum amount of the desired products in the off-gas. The conversion of the hydrocarbon in-gas to acetylene increases with temperature, but as the amount of combustion is increased to obtain the higher temperature, it reduces the hydrocarbon available for conversion

to acetylene, and these two opposite variables must be controlled to obtain optimum results. For a given size and design, this optimum can be correlated with temperature, and the operator must control the amount of air or oxygen supply to maintain this temperature.

The in-gas should have a velocity in the channels of from 30 to 120 pounds per square foot area of the channels. This mass velocity limit applies to all in-gas regardless of its composition. The air injected through the nozzles **33** should have an even higher velocity. To obtain an off-gas containing a substantial proportion of acetylene, the volume of the central space **16** depends upon the composition of the in-gas, the operating temperature, and the mass velocity. We have found that the volume should be from 10% to 40% of the total volume of all channels **14** in both masses **12** and **13**. The ratio of volumes of steam to hydrocarbon in in-gas may be from 1.0 to 6.0. The RH flow must be stopped before the RH end of the LH mass **12** becomes so cooled that partial combustion does not occur therein. The point at which the RH flow must be stopped can be readily determined by a thermocouple near the RH end of the LH mass and the flow can be continued until this temperature falls to, say, 1000° C. which may take several minutes. In practice, the timer is set to stop the RH flow in from one to four minutes after it is started. During this RH flow the LH end of the LH mass is cooled by the flow of in-gas therethrough and the entire RH mass **13** is heated as it cools the off-gas. The RH flow must, however, also be stopped before the off-gas delivered to the pipe **20** reaches a temperature at which acetylene is unstable. In practice this temperature is assumed, for safety, to be 450° C. In some plants it is desirable to inject a small stream of water into the space **17** during RH flow and into the space **15** during LH flow. The vaporization of this water, of course, lowers the temperature of the off-gas.

The RH flow is stopped by closing the valves **58, 60** and **51** and the LH flow is started by opening the valves **56, 61,** and **52**. These valve changes are made automatically and very quickly so that the flow of gases into, through, and out of the furnace is interrupted for less than one second. The LH flow from the pipe **20** to the pipe **18** after the aparatus has operated cyclically for a few minutes is exactly like the RH flow except conditions are reversed. The in-gas picks up heat stored in the RH mass during RH flow and the off-gas delivers heat to the LH mass.

The apparatus can, of course, be used to practice various processes. The preferred process is that above described and this process may be used to convert an in-gas containing any suitable hydrocarbon to an off-gas containing a substantial proportion of acetylene. This process can be applied to the conversion of methane or a gas such as natural gas to an off-gas which contains a substantial amount of acetylene. The process operates very well when applied to the olefines which react to acetylene at lower temperatures than methane.

We claim as our invention:

1. A furnace for subjecting gases to endothermic reactions comprising: a single gas-tight shell having a single heat insulating lining disposed about a single central longitudinal axis; two regenerative masses inside said lining and dividing the space into two end spaces and a central space, the inner ends of said masses defining said central space being substantially parallel and perpendicular to said axis each of said masses having channels passing horizontally therethrough and connecting one of said end spaces with said central space the two masses being placed symmetrically about said axis; a conduit so placed as to either deliver an in-gas to the LH end space or to withdraw an off-gas from said space; a conduit so placed as to either deliver an in-gas to the RH end space or withdraw an off-gas from said space; a series of nozzles so placed as to inject streams of gas into said central space close to the RH end of said LH mass; and a second series of nozzles so placed as to inject streams of gas into said central space close to the LH end of said RH mass, all of said nozzles in each of said series being substantially parallel and in a transverse plane in said central space parallel and close to the said inner end of one of said masses so as to blend said streams of gas from all of the nozzles in each of said series substantially into a single sheet of gas in said plane.

2. A furnace as specified in claim 1 in which some of the nozzles in each of said series of nozzles are directed into said central space from one side of the furnace and the remaining nozzles in that series are directed into said central space from the other side of said furnace.

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