Dec. 28, 1954 J. L. BILLS 2,698,349

ACETYLENE MANUFACTURE

Filed Sept. 9, 1949 2 Sheets-Sheet 1

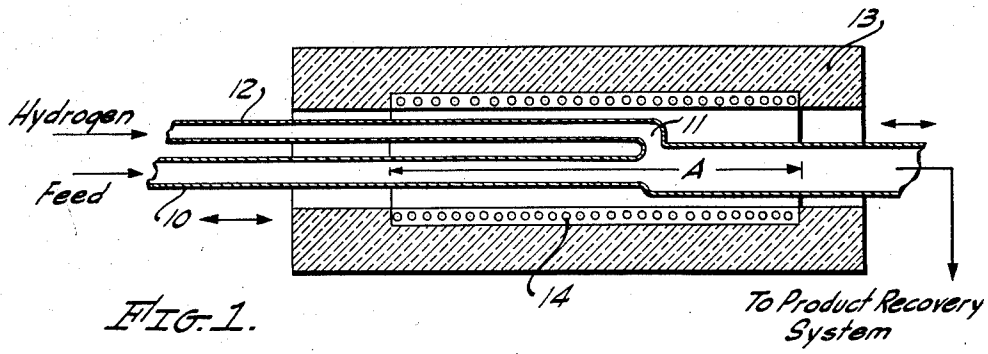


FIG. 1.

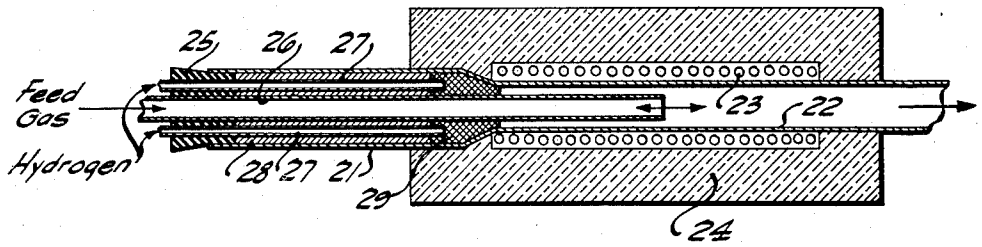


FIG. 2.

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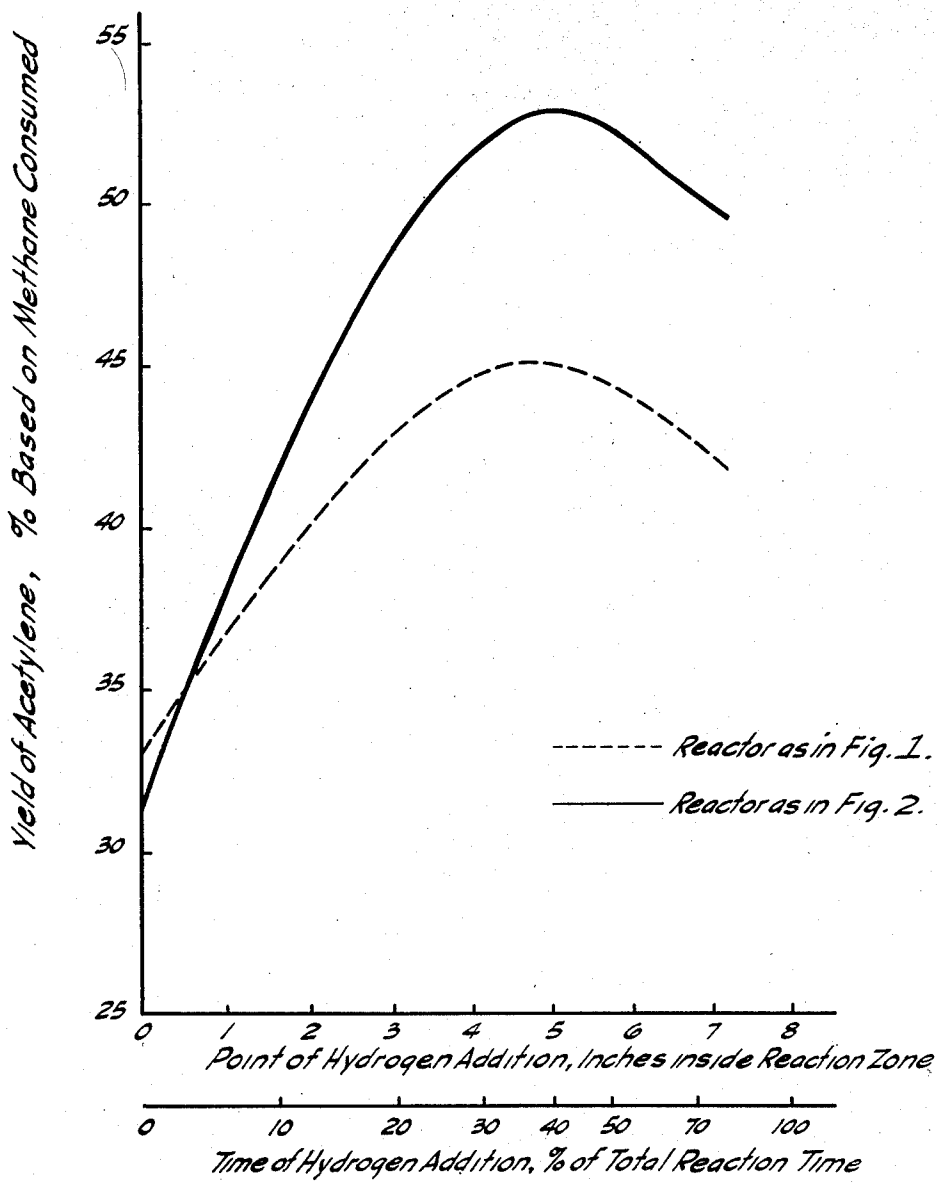


FIG. 3.

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2,698,349

ACETYLENE MANUFACTURE

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Application September 9, 1949, Serial No. 114,719

10 Claims. (Cl. 260—679)

This invention relates to the manufacture of acetylene, and in particular concerns an improved process for obtaining acetylene by the partial oxidation of saturated hydrocarbons.

It is known that acetylene may be obtained by the partial oxidation of hydrocarbons, particularly saturated aliphatic hydrocarbons such as methane, ethane, natural gas, etc., with oxygen or an oxygen-containing gas such as air. Such oxidation, however, does not occur to any significant extent except at elevated temperatures which favor a variety of side reactions, and consequently the yields of acetylene realized are far below theoretical. When air is employed as the oxidizing agent the yield of acetylene is even further reduced as a result of the diluent effect of the large quantity of inert nitrogen present. Accordingly, the best yields of acetylene have been obtained by employing pure oxygen as the oxidizing agent and, even so, have not amounted to more than about 40 per cent, based on the amount of hydrocarbon consumed in the reaction. Such low yield and the high cost of the pure oxygen required, particularly the latter, have up to the present rendered the process commercially impractical despite the availability of large quantities of suitable saturated hydrocarbons in the form of natural gas, waste refinery gases, etc.

It is accordingly an object of the present invention to provide an improved process for the production of acetylene by the partial oxidation of hydrocarbons.

Another object is to provide a process whereby acetylene may be produced from saturated hydrocarbons in yields higher than those attained heretofore.

A further object is to provide a process whereby saturated hydrocarbons are caused to react with air to form acetylene in yields as good or better than those heretofore realized in processes employing relatively pure oxygen as the oxidizing agent.

Other objects will be apparent from the following detailed description of the invention, and various advantages not specifically referred to herein will occur to those skilled in the art upon employment of the invention in practice.

I have now found that the above and related objects may be realized in a process whereby hydrogen is added to a reactant gas mixture comprising a hydrocarbon and oxygen while said mixture is undergoing reaction at a high temperature over a short period of time. More particularly, I have found that acetylene may be produced in relatively high yields by passing a gaseous mixture consisting essentially of a hydrocarbon, particularly a saturated aliphatic hydrocarbon such as methane or natural gas, and oxygen or an oxygen-containing gas through a reaction zone maintained at an acetylene-producing temperature, e. g., between about 1100° and about 1500° C., at such a rate that the time of passage through the reaction zone is between about 0.005 and about 0.02 seconds, and simultaneously adding hydrogen to the reactant gas stream at a point intermediate along the length of the reaction zone. I am aware that it has been proposed to add hydrogen to a similar reactant gas mixture prior to its passage through a high temperature reaction zone, but I have found that such procedure does not attain the advantageous results secured by adding the hydrogen to the reactant mixture during the course of the reaction, i. e., after the reaction has become initiated within the reaction zone. Furthermore, I have found that there is an optimum point for the addition of the hydrogen during the reaction, whereby maximum

yields of acetylene are obtained. The location of such point depends primarily upon the reaction time, i. e., upon the time of passage of the reactant gas mixture through the reaction zone, although other factors are involved to a lesser extent, and accordingly is best expressed in terms of time. In general, highest yields of acetylene are obtained when the hydrogen is added to the reactant gas mixture after the elapse of from about 25 to about 75 per cent of the total reaction time. In most instances such period of time amounts to between about 0.002 and about 0.015 second.

The invention thus consists in a process whereby a reactant gas mixture essentially comprising a hydrocarbon and oxygen is subjected to an acetylene-producing temperature for from about 0.005 to about 0.02 second, during which time hydrogen is added to the mixture, preferably after the elapse of from about 25 to about 75 per cent of the total reaction time. In addition to producing acetylene in yields considerably higher than those realized in previous processes, such procedure is further advantageous in that it completely eliminates the formation of free carbon as one of the products of the reaction. Furthermore, the reactions involved when employing the present procedure are more highly exothermic than those of the previously known processes, and accordingly the heat requirements of the present process are substantially lower. Also, the production of carbon dioxide as a by-product is markedly decreased.

The nature of the invention may be readily understood by reference to the accompanying drawings which form a part of this specification. In said drawings, Figure 1 diagrammatically represents a longitudinal cross-section of a simple apparatus which has been experimentally employed to demonstrate the nature of the invention. Figure 2 represents a longitudinal cross-section of a somewhat different form of apparatus which may be employed in practicing the invention. Figure 3 graphically illustrates the relationship which exists between the yield of acetylene and the point of hydrogen addition employing reactors similar to those illustrated by Figures 1 and 2 under certain operating conditions more fully described hereinafter.

Referring now to Figure 1, the illustrated apparatus consists simply of a quartz tube **10** fitted at point **11** along its length with a side-arm **12**. Beyond point **11**, tube **10** is enlarged to about twice its initial diameter. The tube and side-arm assembly is movably supported in furnace **13** in such manner that it may be moved horizontally with respect thereto. Furnace **13** is provided with electrical heating means, shown as a heating coil **14**, capable of providing a high temperature within tube **10**. The extent of the heating means along the length of tube **10** for practical purposes defines a reaction zone A within tube **10**. A gaseous mixture of hydrocarbon and oxygen or air is introduced into tube **10** and hydrogen is introduced into the side arm **12**. Within tube **10** the reaction mixture passes through reaction zone A within which, at point **11**, it is joined by the stream of hydrogen introduced into side-arm **12**. It will be seen that by moving the tube and side-arm assembly in and out of the furnace **13**, the point at which hydrogen is added to the reactant gas may be varied with respect to the reaction zone A so that the hydrogen may be added to the reactant gas at any time during its passage through the reaction zone. The product gases withdrawn from the enlarged portion of tube **10** are passed to a product recovery system, not shown.

The following example typifies experiments carried out with the apparatus of Figure 1 to demonstrate the effect of adding hydrogen to the reactant gas mixture at different points along the reaction zone.

Example I

The reactor was constructed substantially as shown in Figure 1, the narrow portion of tube **10** and side-arm **12** each having an inside diameter of about ⅛ inch, and the enlarged portion of tube **10** having a diameter of about ¼ inch. The heating element was regulated to maintain a temperature of about 1200° C. along an 8-inch length of tube **10**, thereby providing a reaction zone 8 inches long. The feed gas, consisting of 24.6 per cent

by volume of methane and 75.4 per cent by volume of air, amounting to a mole ratio of methane to oxygen of about 1.63/1, was introduced into the reactor at a rate of 7.94 s. c. f./hr., and hydrogen was introduced into the side-arm at a rate of 3.89 s. c. f./hr. The mole ratio of hydrogen to methane was about 2/1. The reaction products wtihdrawn from the reactor were immediately cooled, passed through a cold trap to knock out condensible products, and the non-condensed gas was thence collected for analysis in a gas sampling system. A series of five experiments was carried out under these conditions, the point of hydrogen addition along the length of the reaction zone being varied in each case by moving the tube and side-arm assembly along the length of the furnace. The data obtained are tabulated as follows:

Experiment	Point of H_2 Addition, Inches [a]	Time of H_2 Addition, Sec. [b]	Time of H_2 Addition, Percent [c]	Acetylene Yield, Percent [d]
1	0.0	0.0	0.0	33.0
2	1.0	0.00066	5.3	37.2
3	3.0	0.00199	19.1	42.8
4	5.0	0.00332	39.7	45.4
5	7.0	0.00464	73.4	42.8

[a] Distance in inches from beginning of the reaction zone.
[b] Time in seconds after entry of the feed gas into the reaction zone.
[c] Percent of total reaction time elapsed before the addition of hydrogen.
[d] Based on amount of methane consumed.

It will be noted that a significant improvement in the yield of acetylene is effected by adding hydrogen to the reactant gas stream within the reaction zone after the elapse of only 5.3 per cent of the total reaction time, amounting to about 0.0007 second, and that the maximum yield of acetylene was obtained by adding the hydrogen after the elapse of about 40 per cent of the total reaction time, amounting to about 0.003 second.

While the apparatus and procedure described in the preceding example satisfactorily illustrate the principle upon which the invention is based, I have found that the yield of acetylene obtained is improved by employing a reactor which is better adapted to promote non-turbulent mixing of the hydrogen and the reactant gas at the point of hydrogen addition, and also to promote laminar flow of the mixture of hydrogen and reactant gas beyond the point of hydrogen addition. Such a reactor is illustrated by Figure 2 of the accompanying drawings, reference to which is now made. Said reactor consists of a quartz tube **21** having a constricted portion **22** which forms an elongated reaction zone. Said reaction zone may be heated to a high temperature by passing an electric current through heating element **23**. An insulating layer **24** covers the heating element **23** and minimizes heat losses to the atmosphere. A stopper **25** closes the open end of tube **21** and serves to support feed gas inlet **26** which is concentrically positioned within tube **21** and extends into the reaction zone **22**. By sliding gas inlet **26** in and out of the stopper **25**, its extent into reaction zone **22** may be varied at will. Stopper **25** also supports hydrogen inlets **27** which extend into tube **21** but not into the reaction zone **22**. A section of tube **21** is packed with an insulating material **28** which serves to protect the stopper **25** from the heat as well as to provide additional support for feed gas inlet **26** and hydrogen inlets **27**. The remainder of tube **21** is packed with nickel gauze **29** which serves as a flame arrestor. The reaction zone **22** leads directly to conventional means, not shown, for cooling and collecting the product gas. The operation of this apparatus will be readily understood when it is considered that that portion of the feed gas inlet **26** which extends into the reaction zone **22** becomes heated to the temperature of the reaction zone and hence comprises part of the reaction zone. Accordingly, the feed gas introduced into inlet **26** begins to react as soon as it reaches the point where the inlet extends into the reaction zone. Reaction in the absence of added hydrogen continues until the reactant gas reaches the end of inlet **26**, whereupon it becomes mixed with hydrogen and the reaction continues in the presence of hydrogen throughout the remaining length of the reaction zone. By sliding the inlet tube in and out of stopper **25**, the time interval between the start of the reaction within the inlet tube and the addition of the hydrogen at the end of the inlet tube may be varied as desired.

The following examples illustrate practice of the invention employing a reactor of the type just described but are not to be construed as limiting the invention:

Example II

The reactor was constructed substantially as shown in Figure 2, except that heating of the reaction zone was accomplished by "Glo-Bar" silicon carbide heating elements rather than by a coil of resistance wire. The reaction zone was about 8 inches long and was maintained at a temperature of about 1200° C. The feed gas inlet was ⅛ inch in diameter, and the constricted portion of tube **22** was ¼ inch in diameter. The feed gas, consisting of about 24.6 per cent by volume of methane and 75.4 per cent by volume of air, was introduced into the feed gas inlet at a rate of 7.94 s. c. f./hr., and hydrogen was introduced into the hydrogen inlets at a rate of 3.89 s. c. f./hr. The mole ratio of hydrogen to methane was about 2/1. The product gas withdrawn from the reactor was shock-cooled, passed through a cold trap, and thence to a conventional gas sampling system. A series of five runs was made under these conditions, the point, and hence the time, at which hydrogen was added being varied by sliding the feed gas inlet in and out of the stopper. The data obtained are tabulated as follows:

Run No.	Point of H_2 Addition, Inches [a]	Time of H_2 Addition, Sec. [b]	Time of H_2 Addition, Percent [c]	Acetylene Yield, Percent [d]
1	0.0	0.00	0.00	31.8
2	1.5	0.0010	9.27	43.0
3	3.0	0.00199	19.1	47.8
4	5.0	0.00332	39.7	53.4
5	7.0	0.00464	73.4	50.4

[a] Distance in inches from beginning of reaction zone.
[b] Time in seconds after entry of the feed gas into the reaction zone.
[c] Percent of total reaction time elapsed before the addition of hydrogen.
[d] Based on amount of methane consumed.

It will be noted that the reaction conditions employed above were the same as those employed in Example I, and that with the exception of Run No. 2 the point of hydrogen addition in the respective runs in each example was likewise the same. Accordingly, Examples I and II are comparable with respect to all variables except the apparatus employed.

The curves shown in Figure 3 of the accompanying drawings are plotted from the data obtained in Examples I and II, and illustrate the relationship between the yield of acetylene and the point of hydrogen addition. Also, a comparison of the two curves indicates the improved results obtained by employing a reactor which promotes non-turbulent mixing of the reactant gas and the added hydrogen. It will further be noted from these curves that the optimum point of hydrogen addition, corresponding to the maximum yield of acetylene, was substantially the same in both sets of experiments.

Example III

The apparatus and procedure employed was the same as that described above in Example II. The operating conditions were as follows:

Feed gas:	
Methane	21.4% by vol.
Air	78.6% by vol.
Rate of feed	5.0 s. c. f./hr.
Rate of hydrogen addition	3.21 s. c. f./hr.
Mole ratio, hydrogen to methane	3/1
Reaction temperature	1200° C.
Total reaction time	0.0112 sec.
Hydrogen added	0.00612 sec. after reactant gas entered reaction zone.
Per cent of total reaction time elapsed before adding hydrogen	59.0

The composition of the non-condensed portion of the product gas was as follows:

	Per cent by volume
Nitrogen	42.6
Hydrogen	45.0
Methane	6.2
Acetylene	3.1
Ethylene	0.5
Carbon monoxide	2.3
Carbon dioxide	0.2
Oxygen	0.1
Total	100.0

The yield of acetylene based on the amount of methane introduced in the feed gas was 39.0 per cent. The yield of acetylene based on the amount of methane consumed in the reaction was 64.0 per cent. The net consumption of the added hydrogen was substantially zero.

In order to demonstrate that the improved yield of acetylene secured in the present process is due to some chemical action of the added hydrogen rather than to some physical effect of an added gas regardless of its chemical identity, there was carried out an experiment wherein nitrogen was substituted for the added hydrogen. The feed gas, consisting of 24.5 per cent by volume of methane and 75.5 per cent by volume of air, was introduced into the reactor at a rate of 5.67 s. c. f./hr. The nitrogen was introduced at a rate of 6.18 s. c. f./hr., corresponding to a nitrogen to methane ratio of 4.4/1, and mixed with the reactant gas in the reaction zone after about 17.7 per cent of the total reaction time had elapsed. A reaction temperature of 1200° C. was employed. The amount of acetylene in the product gas was substantially zero.

Similarly, in order to demonstrate that the improved results attained by the present process are not primarily due merely to the introduction of heat into the reactant gas by the added hydrogen, the apparatus was arranged so that the added hydrogen was preheated to the reaction temperature and then mixed with the reactant gas just as the latter entered the reaction zone. The yield of acetylene, based on the methane consumed, was only 34.2 per cent.

Finally, in order to demonstrate the effect of omitting the added hydrogen entirely, a feed gas containing 21.6 per cent by volume of methane and 78.4 per cent by volume of air was passed through the reaction zone maintained at a temperature of 1200° C. at a rate of 2.5 s. c. f./hr., corresponding to a reaction time of 0.0163 sec. The yield of acetylene, based on the amount of methane consumed, was only 10.8 per cent.

Considering now the essential operating variables in somewhat greater detail, the reactant gas consists essentially of a proportioned mixture of a hydrocarbon and oxygen. The hydrocarbon reactant is preferably a normally gaseous saturated aliphatic hydrocarbon, i. e., methane, ethane, or propane, although liquid saturated aliphatic hydrocarbons of higher molecular weight may be employed if desired. Also, hydrocarbons other than saturated aliphatic hydrocarbons, e. g., olefines, naphthenes, aromatics, etc., may be employed if desired. When employing a liquid hydrocarbon reactant, means should be provided for vaporizing such liquid hydrocarbon prior to its introduction into the reaction zone in admixture with the oxygen reactant. For economic reasons, methane, particularly as it occurs in natural gas, is preferred. The oxygen reactant may be pure oxygen itself, oxygen-enriched air, ordinary air, or any other oxygen-containing gas. As previously stated, air is preferred by reason of its lack of cost, and it is one of the features of the present process that the results obtained employing air are comparable or better than those of previous processes in which pure oxygen was employed. The mole ratio of hydrocarbon to oxygen in the reactant gas mixture may be varied between rather wide limits, but in order to suppress undesirable side reactions the hydrocarbon should be present in an amount not less than that required by theory for reaction with the amount of oxygen present to produce acetylene. When methane or natural gas is employed as the hydrocarbon reactant, the mole ratio of hydrocarbon to oxygen should be greater than about 1.33/1 and is suitably between about 1:33/1 and about 2.0/1, preferably between about 1.5/1 and 1.8/1. When the oxygen reactant is provided in the form of air and the hydrocarbon reactant is methane or natural gas, the reactant gas may comprise from about 21 to about 30 per cent by volume of the hydrocarbon and, correspondingly, from about 79 to about 70 per cent by volume of air.

The amount of hydrogen added to the reactant gas during its passage through the reaction zone may likewise be varied considerably since there is at most only very little net consumption of the added hydrogen in the reaction. Usually, however, it is preferred to add from about 0.5 to about 5 moles of hydrogen per mole of hydrocarbon in the reactant gas. Such added hydrogen is advantageously preheated approximately to the reaction temperature prior to its addition to the reactant gas within the reaction zone. In the reactors shown in Figures 1 and 2, such preheating of the added hydrogen is inherently provided by the design of the apparatus, particularly when the point of hydrogen addition is well within the reaction zone. However, independent means for preheating the hydrogen may be provided if desired.

The reaction temperature and the reaction time are more or less interdependent, shorter reaction times being employed at the higher reaction temperatures and vice versa. Ordinarily, the reaction temperature will be above about 1000° C., usually between about 1100° and about 1500° C., and the reaction time will vary from about 0.005 to about 0.02 second.

As is illustrated by the data obtained in the experiments described in Examples I and II, above, and by the graphical presentation of these data in Figure 3 of the accompanying drawings, a substantial increase in the yield of acetylene may be obtained by adding the hydrogen to the reactant gas within the reaction zone only a very short time after the entry of said gas into said zone. Also, some degree of improvement in acetylene yield is secured when the point of hydrogen addition is substantially at the end of the reaction zone, i. e., substantially at the end of the reaction period. Accordingly, the objects of the invention may be attained to a certain extent by adding the hydrogen to the reactant gas at any time during the course of the reaction, i. e., at any point along the length of the reaction zone. From a practical standpoint, the addition of hydrogen may advantageously be made after the elapse of from about 5 to about 95 per cent of the total reaction time. The optimum time for the addition of the hydrogen, whereby maximum yields of acetylene are attained, depends upon the other operating variables, e. g., the reaction temperature, the reaction time, the feed gas composition, etc. Ordinarily, however, such optimum time will be between about 25 and about 75 per cent of the total reaction time, i. e., highest yields of acetylene are usually attained when the hydrogen is added to the reactant gas mixture within the reaction zone after 25–75 per cent of the total reaction time has elapsed. Under the usual operating conditions, such time amounts to between about 0.002 and about 0.015 second.

The advantage of employing a reactor which promotes non-turbulent mixing of the added hydrogen and the reactant gas is also apparent from the curves of Figure 3. Accordingly, it is preferred that the reactor employed be of such type. Such reactor may provide for the addition of hydrogen to the reactant gas stream in a direction concurrent with the flow of the latter, as is accomplished in the apparatus illustrated by Figure 2. Alternatively, the stream of added hydrogen may be tangentially introduced into the reactant gas stream. Other means of securing non-turbulent mixing of gas streams are known in the art.

As will be apparent to those skilled in the art, many variations with respect to the different operating variables, reactor design, etc., are possible within the herein defined scope of the invention, and various engineering techniques may be applied to the practice of the invention on a large scale. Thus, for example, it may be desired to preheat the feed gas by heat exchange against the hot product gas or by other means. Similarly, various forms of heating equipment may be employed to provide the required high reaction temperature, and the reactor may take various forms adapted to conserve heat as much as possible, and may be constructed of various refractory materials. Likewise, the product gas may be treated in various known ways to separate the different constituents thereof. For example, the unreacted hydrocarbon and hydrogen may be recovered for re-use in the

process by selective adsorption on activated charcoal employing the known "Hypersorption" process. The acetylene may be separated by adsorption on a solid adsorbent, by selective solvent extraction or by a selective chemical reaction such as absorption in aqueous solutions of certain metal salts. Also, the feed gas constituents may be obtained from various sources, and the feed gas may be prepared in various ways. Thus, a suitable feed gas mixture may be obtained according to the process described and claimed in my copending application, Serial No. 125,663 filed November 4, 1949, whereby a hydrocarbon gas is contacted with barium peroxide at an elevated temperature to obtain a gas mixture comprising the hydrocarbon and oxygen, and the barium oxide produced by such treatment is subsequently contacted with air at an elevated temperature to regenerate the peroxide for re-use in the process.

The principle upon which the present invention is based may also be applied to the partial oxidation of hydrocarbons to form olefines, i. e., improved yields of olefines may be obtained by adding hydrogen to a proportioned mixture of hydrocarbon and oxygen undergoing reaction to form olefines at a high temperature over a short period of time in the known manner. Similarly, in the high temperature reaction between a hydrocarbon, oxygen and ammonia to produce hydrogen cyanide, improved yields of the latter may be obtained by adding the ammonia reactant some time after introduction of the hydrocarbon and oxygen into the high temperature reaction zone.

Other modes of applying the principle of my invention may be employed instead of those explained, change being made as regards the materials and apparatus employed, provided the step or steps stated by any of the following claims, or the equivalent of such stated step or steps be employed.

I, therefore, particularly point out and distinctly claim as my invention:

1. A process of producing acetylene which comprises passing a reactant gas mixture containing from about 21 to about 30 per cent by volume of methane and from about 79 to about 70 per cent by volume of air through a reaction zone maintained at an acetylene-producing temperature between about 1100° C. and about 1500° C. at such a rate that the residence time of said reactant gas mixture within said reaction zone is between about 0.005 and about 0.02 second, and simultaneously introducing a gas consisting of hydrogen into said reaction zone at such point along the length thereof and in such manner that said hydrogen is substantially non-turbulently mixed with said reactant gas mixture within said reaction zone after the elapse of from about 25 to about 75 per cent of the total reaction time, said hydrogen having been preheated to a temperature substantially equal to the temperature maintained within said reaction zone prior to its introduction thereinto and sufficient of said hydrogen being introduced into said reaction zone to provide from about 0.5 to about 5 moles of hydrogen per mole of methane present in said reaction zone.

2. In a process for the production of acetylene wherein a reactant gas mixture essentially comprising hydrocarbon and oxygen is continuously passed through an elongated reaction zone maintained at an acetylene-producing temperature above about 1000° C. and within which an acetylene-producing reaction occurs, the step which comprises continuously admixing a gas consisting of hydrogen with the gases undergoing reaction within said zone after the start but before the completion of said reaction by introducing said stream of hydrogen into said reaction zone at a point intermediate along the length thereof.

3. In a process for the production of acetylene wherein a reactant gas mixture essentially comprising a saturated aliphatic hydrocarbon and oxygen is continuously passed through an elongated reaction zone maintained at an acetylene-producing temperature above about 1000° C. and within which an acetylene-producing reaction occurs, said hydrocarbon being present in the reactant gas mixture in an amount at least equal to that theoretically required for reaction with the amount of oxygen present to produce acetylene, and the rate of passage of said gas mixture through said reaction zone being such that the residence time of the gas within the reaction zone is between about 0.005 and about 0.02 second, the step which comprises continuously admixing a gas consisting of hydrogen with the gases undergoing reaction within said zone after the start but before the completion of said reaction by introducing said stream of hydrogen into said reaction zone at a point intermediate along the length thereof.

4. A process according to claim 3 in which the reactant gas mixture essentially comprises methane and oxygen in a mole ratio at least equal to about 1.33/1 and the reaction zone is maintained at an acetylene-producing temperature between about 1100° C. and about 1500° C.

5. A process according to claim 3 wherein the hydrogen is preheated to a temperature substantially equal to that maintained within the reaction zone prior to its introduction thereinto.

6. A process according to claim 3 wherein the amount of hydrogen introduced into the reaction zone is sufficient to provide from about 0.5 to about 5 moles of hydrogen per mole of methane in the reactant gas.

7. A process according to claim 3 wherein the reactant gas mixture comprises from about 21 to about 30 per cent by volume of methane and from about 79 to about 70 per cent by volume of air, and the reaction zone is maintained at a temperature between about 1100° C. and about 1500° C.

8. A process according to claim 7 wherein the hydrogen is preheated to a temperature substantially equal to that maintained within the reaction zone prior to its introduction thereinto.

9. A process according to claim 7 wherein the amount of hydrogen introduced into said reaction zone is sufficient to provide from about 0.5 to about 5 moles of hydrogen per mole of methane in the reactant gas.

10. A process according to claim 7 wherein the hydrogen is introduced into the reaction zone at such point that it is caused to be admixed with the gases undergoing reaction within said zone after the elapse of from about 25 to about 75 per cent of the total reaction time.

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