Nov. 1, 1960

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2,958,716

PROCESS FOR USING SHOCK WAVES TO PRODUCE ACETYLENE

Filed Nov. 20, 1957

2 Sheets-Sheet 1

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PROCESS FOR USING SHOCK WAVES TO PRODUCE ACETYLENE

Fig. 3.

Fig. 4.

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This invention relates to a novel process for producing acetylene from hydrocarbons by the use of shock waves. This process is a continuation-in-part of U.S. application No. 558,425 by the same applicants filed October 4, 1955 and now abandoned.

It is known that hydrocarbons, including methane as well as higher aliphatic and aromatic hydrocarbons, will yield acetylene when subjected to partial combustion or thermal cracking processes. In thermal cracking processes the necessary heat for the reaction is supplied to a charge by direct heating, and in partial combustion processes, the necessary heat for the reaction is supplied by the combustion part of the charge itself. In either case, high temperature is required to cause the reaction desired in the formation of acetylene, and rapid cooling is also required in order to avoid decomposition and polymerization of the acetylene formed at the higher temperatures and thereby permit recovery of the acetylene itself.

It has now been found that the reaction conditions required for making acetylene from hydrocarbons can be met by subjecting the hydrocarbons to a shock wave. One way of obtaining such a shock wave is by means of a detonation. The term "detonation" is meant a very rapid combustion in which the flame front moves at velocities higher than the speed of sound in the unburnt gases. The rate of flame propagation is thus far greater in a detonation than in a deflagration, which is a combustion in which the velocity of flame propagation does not exceed the speed of sound in the unburnt gases. According to Wilhelm Jost's "Explosion and Combustion Processes in Gases," McGraw-Hill Book Company, Inc., New York (1946), pages 160 to 210, all of which are devoted to detonations, the velocity of the flame front in detonations thus far investigated is from 1 to 4 kilometers per second (about 3,280 to 13,120 feet per second), as compared to, for instance, 50 feet per second for a typical explosion.

The flame of a detonation moves into unburned gases with a velocity that is supersonic instead of subsonic, and it is initiated by and remains associated with a shock front that is characterized by almost instantaneous rise and very rapid fall of temperature and pressure. Once established in an elongated chamber, the detonation wave travels at a constant velocity (Lewis and Von Elbe, "Combustion, Flames and Explosions," Academic Press, Inc., 1951). We have found that these unique properties of the detonation phenomenon render them especially useful in a process for producing acetylene.

A method of producing shock waves suitable for the production of acetylene from hydrocarbons is by the use of a shock tube. A membrane is placed in the tube defining two chambers therein, the ends of the tube being closed. Gas pressure is built up on one side of the membrane while the other side is evacuated and partially filled with a hydrocarbon make gas. When the pressure differential between the two chambers is sufficient, the membrane is ruptured either by pressure or mechanically. A supersonic shock wave is thereby set up as the high pressure gas expands into the low pressure gas. This wave travels along the tube at sonic speed setting up associated rarefaction and compression zones having associated high and low temperature areas, respectively, in the zones sufficient to cause cracking of the make gas to acetylene. This process will be more fully set forth below with reference to drawings and examples.

It is a primary object of this invention to provide a process for utilizing shock waves to make acetylene from other hydrocarbons. It is a further object to provide such a process wherein the shock waves are derived from either a shock tube reaction or from a detonation gun.

According to one embodiment of this invention, acetylene can be produced from other hydrocarbons by passing the shock wave resulting from a detonation into a confined charge of the hydrocarbon to be cracked. In order to maintain the well-defined interface between the detonation charge and the cracking charge prior to the detonation, the charges may be physically separated by a diaphragm or membrane which is easily ruptured by and does not objectionably interfere with the passage of the shock wave into the cracking stock. It has been found that a membrane is not necessary to provide separation of the detonation charge and the cracking charge. Alternatively, an inert gas barrier may be placed between the two charges or both charges may be flowed through the gas tube opposite to each other and vented at the center thus forming a dynamic interface between the two, thereby preventing substantial mixing of the detonation and cracking gas charges before the detonation. Both of these latter methods lend themselves more nearly to continuous production as it is not necessary to physically replace the membrane after each detonation. The cracking charge may be either detonable or non-detonable when exposed to the shock wave from another detonation. The cracking charge may be made up entirely of the hydrocarbon to be cracked or it may be mixed with an oxidant such as oxygen. Some hydrocarbon-oxygen mixtures are not detonable unless exposed to a high energy source such as that provided by the shock wave of another detonation. Where no detonation occurs in the cracking charge, the effective penetration of the detonation shock wave into the charge seems to depend on the intensity of the detonation charge. Analyses indicated that a major part of the acetylene formed in these experiments was found near the boundary formed by the separating membrane, the area of most effective penetration. Where the components of the cracking stock are in such proportions to result in a mixture detonable by the shock wave resulting from detonation of the detonation charge, impingement of the shock wave thereagainst results in a continuation of the propagation of the detonation in the cracking stock.

These and other objects, features and advantages will become apparent from the following detailed description of the accompanying drawings in which:

Fig. 1 is a schematic longitudinal sectional view of one form of reaction apparatus in which the process of the present invention may be practiced;

Fig. 2 is a view of a longitudinal section through another form of reaction apparatus capable of carrying out the principles of the present invention;

Fig. 3 is a longitudinal sectional view of a shock tube adapted for carrying out the method of the instant invention; and

Fig. 4 is a longitudinal sectional view of a modified form of detonation gun in which a membrane is not utilized to separate the detonation and make gases.

Referring more particularly to the drawings, Fig. 1...
shows a reaction apparatus 10 which comprises one
chamber 12 for forming and detonating a detonation
charge and another chamber 14 for receiving a cracking
charge. Detonation chamber 12 is in the form of an
elongated member 52 which is connected at one end with the cracking chamber through a
flanged union 16 which supports a membrane 18 extend-
ing transversely across the opening between the two
chambers. The membrane may, for example, be made of cello-
phone or thin kraft paper. A combustible gas, such as
hydrogen, is supplied through pipes 20, and an oxidizing
gas, such as air, is supplied through a pipe 22, to a mixing
chamber 24 in such proportions as to form a detonable
gaseous charge mixture which passes through a short
connecting pipe 26 into an ignition chamber 28 provided
with spark plug 30. The end of the cracking chamber 14
remote from the detonation chamber is closed by a cap
32 formed with an opening for receiving a fuel supply
line 34 through which the cracking stock is introduced
into cracking chamber. A suitable valve 36 is disposed
in supply line 34 to control the feed into the cracking
chamber. Chambers 12 and 14 are respectively provided
with vents 38 and 40 located on opposite sides of the
separating membrane 18. Acetylene product is with-
drawn through vent 40.

It is to be understood that the term make or cracking
charge refers to the hydrocarbon to be operated on by the
reaction vessel several times as long as it is wide and is
the shock tube while the term detonation charge refers to the gas in
which the detonation is initiated.

The detonation chamber may, for example, be filled
with a charge comprising a stoichiometric mixture of hy-
drogen and oxygen, and the cracking chamber may be
filled with a gaseous hydrocarbon with or without added
oxygen. The desired reaction is effected by igniting
the detonation charge in chamber 12. The resulting shock
wave ruptures the separating membrane, and passes into
the cracking stock.

In the modification of Fig. 2, a shorter and conically-
shaped detonation chamber 50 is employed. Use of this
shape is economical for it permits a reduction in the
amount of fuel required to produce a detonation wave
having a diameter substantially equal to the inside diam-
eter of the cracking chamber 52, i.e. the cone shape of the
detonation chamber reduces the volume of the chamber
without reduction in the diameter of the detonation wave
available for penetrating the cracking charge.

It is important to present a detonation wave over the
entire cross section of the cracking chamber in order to
subject a maximum amount of cracking stock to the
detonation wave, for the effective penetration of the
detonation may not extend throughout the length of the
cracking chamber. A helix 54 of copper wire is placed
near the ignition end of the detonation chamber 50 in
order to accelerate the formation of detonations in the
shorter detonation chamber. The detonating mixture is
introduced through a connection 56 into the detonation
chamber 50 which is provided with a spark plug 58 near
the entrance of the chamber for ignition of the charge.

The cracking chamber 52 is a short narrow chamber
having a diameter substantially equal to the diameter of the
detonation chamber at the end which is connected to it.
It is normally separated from the latter during the
charging period by a diaphragm 60 which is rupturable by
detonation. Diaphragm 60 is supported by the union
62 connecting the cracking and detonation chambers.
Cracking stock is fed into the cracking chamber through
an inlet connection 64 at a rate controlled by valve 66.
Chambers 50 and 52 are provided respectively with vents
68 and 70, which are disposed on opposite sides of the
diaphragm 60. Product acetylene is removed through
vent 70.

There is wide latitude of choice in the dimensions of the
detonation chamber, provided a detonation can be
formed and directed. In the form of apparatus of Fig. 1,
detonations can be successfully produced with lengths
from fifteen to one hundred and twenty inches when using
a one-inch inside diameter chamber. Various sizes of
cracking chambers ranging from a one-inch diameter
chamber one inch long to fourteen inches long have been
successfully used. Acetylene yields achieved with crack-
ing chambers two inches in length were as good as those
achieved with the longer chambers. In the apparatus of
Fig. 2, conical detonation chambers of six and fifteen
inches in length were used successfully, both chambers
having inside diameters of 0.364 inch at one end and 1.05
inches at its other end. Cracking chambers were
similar to these mentioned above in connection with Fig.
1. The thickness of the separating membrane may also
be varied so long as it does not objectionably interfere
with the passage of the detonation. Cellophane was used
successfully and kraft paper ranging in thickness from
.0004 to .0016 inch was employed. Recovery decreased
as the thickness of the diaphragm increased.

The detonation wave may be generated in a wide
variety of fluid mixtures capable of producing deton-
ations. A stoichiometric mixture of hydrogen and oxygen
has been found very suitable, for it produces a strong
detonation and condensible products. As for cracking
stock, either a gas or vaporized or finely dispersed
liquid hydrocarbon with or without added oxygen can
be used. Suitable hydrocarbons, which have been used
include methane, ethane, propane and n-pentane. Good
yields have been obtained in using oxygen-hydrocarbon
mixtures that are rich in hydrocarbon.

Table I shows the results of several runs using various
cracking stocks in the detonation and cracking apparatus
of Fig. 1 of the drawings. In each instance, the detonat-
ing mixture used was made up of 67% hydrogen and 33% oxygen.

<table>
<thead>
<tr>
<th>Test No.</th>
<th>Cracking Stock</th>
<th>Cracking Chamber</th>
<th>Membrane</th>
<th>Acetylene Yield, cc</th>
<th>Cracking Stock Converted to Cals (percent)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 (182).</td>
<td>Methane... 1&quot; dia. 2&quot; lg. trenlined (22.6 cc. Vol.)</td>
<td>0.0004 Kraft paper...</td>
<td>0.661</td>
<td>7.28</td>
<td></td>
</tr>
<tr>
<td>2 (49).</td>
<td>do... 1&quot; dia. (18 cc. Vol.)</td>
<td>Cellophane...</td>
<td>0.306</td>
<td>2.22</td>
<td></td>
</tr>
<tr>
<td>3 (145).</td>
<td>do... 1&quot; dia. 1&quot; lg. (11.6 cc. Vol.)</td>
<td>0.0004 Kraft paper...</td>
<td>0.206</td>
<td>2.29</td>
<td></td>
</tr>
<tr>
<td>4 (130).</td>
<td>do... 1&quot; dia. 2&quot; lg. (25.6 cc. Vol.)</td>
<td>...</td>
<td>1.83</td>
<td>7.12</td>
<td></td>
</tr>
<tr>
<td>5 (150).</td>
<td>Ethane... 1&quot; dia. 6&quot; lg. (84.5 cc. Vol.)</td>
<td>...</td>
<td>1.85</td>
<td>2.30</td>
<td></td>
</tr>
<tr>
<td>6 (128).</td>
<td>n-pentane... 1&quot; dia. 2&quot; lg. (35.6 cc. Vol.)</td>
<td>...</td>
<td>1.85</td>
<td>7.22</td>
<td></td>
</tr>
<tr>
<td>7 (160).</td>
<td>Propane... 1&quot; dia. 2&quot; lg. (25.6 cc. Vol.)</td>
<td>...</td>
<td>1.95</td>
<td>7.6</td>
<td></td>
</tr>
</tbody>
</table>
It will be seen that the yield obtained in a cracking chamber having a two-inch long diameter in run 4 was greater than the yield obtained in a cracking chamber having six-inch long diameter, as in run 5, when employing the same cracking stock. This seems to indicate that the reaction takes place principally in the portion of the cracking chamber near the diaphragm in the area where penetration is most effective.

Table II shows the results of several runs made with conically shaped detonation chambers, the cracking stock being ethane and the detonating mixture being 67% hydrogen and 33% oxygen in each instance. The cracking chamber used for each run was a one-inch diameter pipe two inches long, and having a volume of 25.6 cc.

<table>
<thead>
<tr>
<th>Test No.</th>
<th>Cracking Stock</th>
<th>Detonation Chamber</th>
<th>Membrane</th>
<th>Acetylene Yield (cc)</th>
<th>Percent Hydrocarbon converted</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>8 (176)</td>
<td>Ethane...</td>
<td>Fig. 2, type, 15° cone, 0.634 to 0.150 dia.</td>
<td>0.0004 Kraft paper...</td>
<td>1.73</td>
<td>6.83</td>
<td></td>
</tr>
<tr>
<td>9 (176)</td>
<td>...do...</td>
<td>Fig. 2, type, 6° cone, 0.634 to 0.150 dia.</td>
<td>...do...</td>
<td>1.22</td>
<td>5.18</td>
<td></td>
</tr>
<tr>
<td>10 (180)</td>
<td>...do...</td>
<td>Fig. 2, type, 6° cone, 0.634 to 1.50 dia.</td>
<td>...do...</td>
<td>2.10</td>
<td>8.27</td>
<td></td>
</tr>
<tr>
<td>11 (280)</td>
<td>...do...</td>
<td>Fig. 2, type, 5° cone, 0.374 to 0.100 dia.</td>
<td>0.0015 Kraft paper...</td>
<td>1.16</td>
<td>4.97</td>
<td>Do.</td>
</tr>
<tr>
<td>12 (280)</td>
<td>...do...</td>
<td>Fig. 2, type, 5° cone, 0.396° to 0.100 dia.</td>
<td>0.0006 Kraft paper...</td>
<td>1.07</td>
<td>6.07</td>
<td>Do.</td>
</tr>
</tbody>
</table>

It will be seen that the use of the two-inch coil to reduce predetonation length in the six-inch conical detonation chamber produced a marked increase in the recovery of acetylene. The effect of diaphragm thickness is also illustrated in the last three runs.

Table III shows the results of several runs made with cracking stocks having added oxygen. Apparatus of the type shown in Fig. 2 was employed, the cracking chamber being one inch in diameter, two inches long and 25.6 cc. in volume and the separating membrane comprising 0.0004 inch thick Kraft paper. In each instance the detonation mixture was 67% hydrogen and 33% oxygen.

<table>
<thead>
<tr>
<th>Test No.</th>
<th>Cracking Stock</th>
<th>Acetylene Basis</th>
<th>Conversion to Carbon</th>
<th>Charge Pressure in Both Chambers</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>18 (190)</td>
<td>52% CH₄ 48% O₂</td>
<td>1.51</td>
<td>Atmospheric</td>
<td>10 p.s.i.g</td>
<td></td>
</tr>
<tr>
<td>14 (181)</td>
<td>57% CH₄ 43% O₂</td>
<td>1.20</td>
<td>10.7</td>
<td>7.95</td>
<td></td>
</tr>
<tr>
<td>16 (181)</td>
<td>46% CH₄ 54% O₂</td>
<td>1.22</td>
<td>10.7</td>
<td>7.95</td>
<td></td>
</tr>
<tr>
<td>18 (201)</td>
<td>56% CH₄ 44% O₂</td>
<td>1.22</td>
<td>10.7</td>
<td>7.95</td>
<td></td>
</tr>
<tr>
<td>17 (201)</td>
<td>55% CH₄ 45% O₂</td>
<td>0.69</td>
<td>3.0</td>
<td>2.0</td>
<td></td>
</tr>
<tr>
<td>16 (203)</td>
<td>57% CH₄ 43% O₂</td>
<td>1.23</td>
<td>10.7</td>
<td>7.95</td>
<td></td>
</tr>
<tr>
<td>19 (209)</td>
<td>57% CH₄ 43% O₂</td>
<td>0.68</td>
<td>3.0</td>
<td>2.0</td>
<td></td>
</tr>
<tr>
<td>20 (209)</td>
<td>67% CH₄ 33% O₂</td>
<td>1.26</td>
<td>10.7</td>
<td>7.95</td>
<td></td>
</tr>
</tbody>
</table>

It has also been found that a barrier is not necessary when a detonation is used as the shock source. In a detonation zone adjacent to a reaction zone, a detonation wave can be initiated which travels toward the reaction zone. This detonation wave is then converted to a shock wave upon entering the reaction zone containing a gas mixture incapable of sustaining a detonation. The shock wave then travels at diminishing strength through the non-detonable medium.

Equipment to carry out this modification of the invention is shown in Figure 4. The reaction vessel consists of a detonation chamber 73 and a reaction chamber 74. A mixture of acetylene and oxygen enters chamber 73 through line 75 and valve 76 and flows out of line 77 and valve 78. A hydrocarbon reactant simultaneously enters chamber 74 through line 79 and valve 80 and flows out of line 77 and valve 78. When chambers 73 and 74 are thus filled valves 76, 78 and 80 are closed and spark igniter 83 is activated to set off a detonation in chamber 73. Product gases are removed from chamber 74 through line 85 and valve 86.

The following examples describe acetylene production by this process using no barrier between the source of shock waves and hydrocarbon.

**Example I**

**ACETYLENE FROM METHANE BY DETONATION-PRODUCED SHOCK WAVES**

A 10-foot length of ½-in.-I.D. steel tubing was connected in a straight line with a 10-foot length of ½-in.-I.D. steel tubing by a ½-in.-I.D. TH whose side outlet led to a valve. The outer end of the ½-in. tube was closed with a fitting bearing a valve and an ignition plug. The outer end of the ½-in. tube was closed with a valve. A gas mixture (67% hydrogen + 33% oxygen by volume) known to develop detonation under the conditions of the experiment was put into the ½-in. tube and a methane-containing gas mixture (67% methane + 33% oxygen) known to be incapable of supporting detonation was put into the ½-in. tube by admitting streams of these compositions through the valves at the ends of the ½-in. and ½-in. tubes, respectively, and allowing them to escape through the valve at the central T. When the gas flows were steady, the inlet valves were closed at the same time; then, in quick succession (to minimize mixing), the outlet valve was closed and the ignition circuit was completed. A clearly audible ping was heard immediately. Analysis by mass spectrometer of the gas in the system after the experiment indicated an acetylene concentration of 0.38% by volume; this corresponds to 0.44 cm³ at room pressure and temperature.

**Example II**

**ACETYLENE FROM METHANE BY DETONATION-PRODUCED SHOCK WAVES**

The apparatus was the same as that used in Example I, above, except that the tubes joined by the ½-in.-bore TH were each 5 feet in length and ½-in. I.D. A mixture of 67% hydrogen and 33% oxygen by volume was put into the ignition end of the system and methane was put into the other end, all at atmospheric pressure, in the way described for Example I, above. Again, a ping was heard on ignition. Analysis by mass spectrometer of the gas in the system after the experiment indicated an acetylene concentration of 0.17% by volume; this corresponds to 0.31 cm³ at room pressure and temperature.

**Example III**

**ACETYLENE FROM METHANE BY DETONATION-PRODUCED SHOCK WAVES**

The apparatus was the same as that used in Example II, above, except that a pressure transducer was present in the fitting at the end of the apparatus opposite the...
ignition end and recording instruments were arranged to record its output. A mixture of 67% hydrogen and 33% oxygen by volume was put into the ignition end of the system and a mixture of 67% methane and 33% oxygen was put into the opposite end in the way described for Example I. A plug was heard on ignition and the record of the output of the present transducer showed that a shock wave had struck the transducer. Analysis by mass spectrometer of the gas in the system after the experiment indicated an acetylene concentration of 0.20% by volume.

A further method of operating the detonation gun shown in Fig. 4 is to flow an inert gas simultaneously into the gun barrel with the make gas and detonation gas. This is done through the valve 78 and line 77. It has been found that satisfactory separation of the detonation and make gases is thus accomplished.

The apparatus used for the shock tube embodiment is shown in Figure 3. It consists of a compression chamber 91 and an expansion chamber 92 separated by a barrier 93, usually of cellophane. The gas used to generate the shock wave is introduced into the compression chamber 91 through line 94 and valve 95 until the desired contained pressure is obtained. The expansion chamber 92 is evacuated by means of a vacuum pump connected through line 96 and valve 97. Hydrocarbon gas or mixture is introduced to expansion chamber 92 through line 98 and valve 99 until the desired pressure is obtained in the expansion chamber 92. Usually the pressure in compression chamber 91 is considerably greater than that in the expansion chamber 92. The barrier 93 is broken either by means of excessive pressure in chamber 91 or by action of a plunger 100. On bursting of the diaphragm, the gas in the compression chamber starts expanding into the expansion chamber. A centered rarefaction wave travels from the diaphragm location back into the compression chamber at sonic speed toward the end of the compression chamber where it reflects. The gas expanding through this rarefaction wave is cooled adiabatically and is accelerated to supersonic speeds. The cooled gas expanding into the expansion chamber adiabatically compresses the gas originally in the chamber, heats it and imparts to it a velocity equal to its own. When this compression wave generated in the low pressure gas is traveling at a supersonic speed relative to that gas, a shock wave is formed. As the shock wave travels through the expansion chamber, it heats up the hydrocarbon molecules. Additional high temperatures are attained at the "outlet" end of the expansion chamber. At this point the shock wave is reflected causing a recompression of the end gases. These gases are then cooled by the reflected rarefaction wave as well as by the on-rushing expanding gases. The product gases are then removed through line 101 and valve 102.

It has been calculated that a shock wave produced by the sudden expansion of gas at 1750 mm. mercury pressure into gas at 5 mm. mercury pressure will have a gas temperature behind the shock front of 5260° C., followed by a zone at -94° C. The temperature of the gases compressed by the reflected shock wave will be 10,380° C. These high temperatures followed by expansion cooling are quite suitable for conversion of hydrocarbons to acetylene.

The following examples describe shock tube production of acetylene.

Example I

ACETYLENE FROM METHANE IN THE SHOCK TUBE

A mixture of 23.3% methane by volume and 76.7% argon was admitted to a 10-ft. section of a shock tube. The argon was used to intensify the shock wave. Hydrogen at above atmospheric pressure was charged into a 4-foot section of the tube, separated by a cellophane diaphragm from the methane and argon. When the diaphragm burst due to the gas pressure, a shock wave formed in the tube which produced an audible report and an orange flash of light. The gas remaining in the shock tube contained 0.34 volume percent acetylene based on a mass spectrometer analysis.

Example II

PYROLYSIS OF BENZENE IN THE SHOCK TUBE

A shock wave was sent through a mixture of 20 volume-percent benzene vapor in argon at one mm. of mercury abs. initial pressure. The shock wave was produced by bursting a cellophane diaphragm separating a 4-foot section of shock tube containing hydrogen at 20 p.s.i.g. (1750 mm. Hg) from a 10-foot section of shock tube containing the benzene vapor mixture. An analysis of the product by mass spectrometer showed that 13% of the benzene was converted to acetylene (on a carbon basis).

Example III

PYROLYSIS OF METHANE IN THE SHOCK TUBE

A shock wave was produced by releasing hydrogen at 20 p.s.i.g. (1750 mm. Hg) into a 50:50 volume percent mixture of methane and xenon by the bursting of a cellophane diaphragm separating the two chambers of a shock tube. The methane was 24.6% converted into acetylene, based on a carbon balance taken from the mass spectrometer analysis of the product.

Example IV

PRODUCTION OF ACETYLENE FROM METHANE AND OXYGEN IN THE SHOCK TUBE

Helium at 1750 mm. abs. pressure was released into a mixture of 75 mole-percent argon, 8.3% oxygen, and 16.7% methane by bursting a cellophane diaphragm separating the two stages of a shock tube. The initial downstream pressure was 12 mm. of mercury. The shock wave which resulted initiated a reaction which was luminous for less than a millisecond. An analysis of the dry gas product, exclusive of helium and argon, based upon a mass spectrometer analysis, is as follows:

<table>
<thead>
<tr>
<th>Volume-percent</th>
<th>CO₂</th>
<th>O₂</th>
<th>CO</th>
<th>C₂H₆</th>
<th>C₂H₄</th>
<th>C₃H₆</th>
<th>C₃H₄</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>2.4</td>
<td>3.5</td>
<td>14.0</td>
<td>0.4</td>
<td>9.7</td>
<td>14.2</td>
<td>54.8</td>
</tr>
<tr>
<td></td>
<td>100.0</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

This is a 45% conversion of the methane to acetylene, based on a carbon balance.

Example V

PRODUCTION OF ACETYLENE FROM METHANE AND OXYGEN IN THE SHOCK TUBE

Acetylene was produced from a mixture of methane, oxygen, and argon in a shock tube as follows: A static pressure of 20 p.s.i.g. (1750 mm. of mercury) of hydrogen was built up in a 4-foot length of the shock tube behind a cellophane diaphragm. A 10-foot section of the shock tube downstream from the diaphragm was evacuated and then filled to 5.8 mm. Hg pressure with a mixture of 53.5% argon, 43.5% methane, and 3% volume-percent oxygen. When the diaphragm was punctured, a bright flash was observed at the windowed section of the tube farthest from the diaphragm. An oscilloscopic trace of the signal from a photomultiplier cell showed that the light flash had dropped to below 30% of maximum intensity after one-half of a millisecond (0.0005 sec.). A sample of the product gas, analyzed by mass spectrometer, showed 14.65 moles of
acetylene per 100 moles of argon. A complete analysis, based on 100 moles of argon is:

<table>
<thead>
<tr>
<th></th>
<th>Moles</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{CO}_2$</td>
<td>4.6</td>
</tr>
<tr>
<td>$\text{C}_2\text{H}_2$</td>
<td>0.54</td>
</tr>
<tr>
<td>$\text{C}_2\text{H}_4$</td>
<td>14.65</td>
</tr>
<tr>
<td>$\text{H}_2\text{O}$</td>
<td>5.44</td>
</tr>
<tr>
<td>$\text{CH}_4$</td>
<td>31.88</td>
</tr>
<tr>
<td>$\text{CO}$</td>
<td>43.50</td>
</tr>
<tr>
<td>$\text{H}_2$</td>
<td>200.00</td>
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<tr>
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**Example VI**

**PRODUCTION OF ACETYLENE FROM METHANE AND OXYGEN IN THE SHOCK TUBE**

Hydrogen at 20 p.s.i.g. was released into a mixture of 34.6% $\text{O}_2$ and 63.4% (by volume) methane at 2.7 mm. of mercury absolute, when a cellophane diaphragm separating them was burst. The shock wave which was produced caused an orange flash of light. A sample of the product gas contained 12% acetylene.

**Example VII**

**PRODUCTION OF ACETYLENE FROM METHANE AND CHLORINE IN THE SHOCK TUBE**

The first stage of the shock tube containing helium gas at 1750 mm. of Hg abs. was separated from a low pressure (1.5 mm. Hg) equimolar mixture of methane and chlorine by a cellophane diaphragm. A shock produced by breaking the diaphragm produced a bright visible flash in the windowed downstream section of the tube. A mass spectrometer analysis of the products showed that 46 mole-percent of the methane was converted into acetylene while 19.5% of the methane remained unchanged.

There has thus been disclosed a novel method for producing acetylene from other hydrocarbons by the use of shock waves. If a detonation gun is used as the source of the shock wave, the detonable gas and the make gas may be separated by a membrane, an inert gas barrier, or by a dynamic interface as when the two gases are vented at their junction. If a shock tube is used as a source of the shock wave, the pressure or shock wave generating gas and the make gas must be separated by a membrane; however, this membrane may be ruptured by either pressure differential or by a mechanical device as disclosed. However, regardless of the source of the shock wave in the make gas, the broad principles of the reaction are essentially the same, i.e., a compression with associated high temperature followed immediately by a rarefaction with associated low temperatures passing through the make gas at supersonic speeds. While specific apparatus capable of performing the method has been disclosed, it is to be understood that other apparatus could equally well be utilized to achieve the desired results. No limitations other than those specifically set forth in the claims are intended.

What is claimed is:

1. A process for transforming hydrocarbons to acetylene by use of shock waves initiated by a detonation comprising the steps of introducing a detonation charge into one end of an elongated reaction space to establish a detonation charge zone, introducing a detonable fluid make charge containing a hydrocarbon to be transformed into another portion of the reaction space, interposing a separating medium between said detonation charge and said fluid make charge, igniting the detonation charge, impinging the resulting detonation wave against the fluid make charge whereby the make charge is caused to detonate and transform a portion of the hydrocarbon of the fluid make charge into acetylene.

2. A process as defined in claim 1, in which said separating medium is a diaphragm rupturable by the detonation wave of said detonation charge.

3. A process for forming acetylene from other hydrocarbons by use of detonations, said process comprising introducing a detonation charge into one portion of a reaction space having a configuration capable of sustaining a detonation, introducing a fluid make charge containing a hydrocarbon other than acetylene into another portion of said reaction space interposing a separating medium between said charges, igniting the detonation charge, and impinging the resulting detonation wave against the make charge, said detonation wave penetrating through said make charge and causing a rapid rise and fall of the temperature and pressure to form acetylene from the hydrocarbon of the make charge.

4. A process as defined in claim 3, wherein said separating medium comprises a diaphragm rupturable by the detonation wave.

5. A process as defined in claim 3, wherein said hydrocarbon is a saturated hydrocarbon.

6. A process as defined in claim 3, wherein said make charge is a mixture comprising ethane and oxygen.

7. A process as defined in claim 3, wherein said make charge is a mixture comprising methane and oxygen.

8. A process as defined in claim 3, wherein said detonation charge is a stoichiometric mixture of hydrogen and oxygen.

9. A process as defined in claim 3, wherein said make charge is a non-detonating hydrocarbon.

10. A process as defined in claim 3, wherein said make charge is made up entirely of a hydrocarbon from the group consisting of methane, ethane, pentane and propane.

11. The process defined by claim 3, wherein the separating medium is achieved by passing an inert barrier gas into the reaction space at a point between the detonation charge and the make charge.

12. The process defined by claim 3, wherein the separating medium is the interface between the detonation charge and the make charge achieved by venting the detonation charge and the make charge at a point approximately midway between their points of introduction in the reaction space.

13. A process for producing acetylene from other hydrocarbons by the use of a shock wave comprising the steps of introducing a make gas into one end of a closed elongated reaction chamber, introducing a second gas into the other end of said reaction chamber, separating the two gases with a thin diaphragm, increasing the pressure of the second gas until a pressure differential exists across the diaphragm capable of initiating a shock wave in the make gas, rupturing the diaphragm, and removing the reaction products.

14. The process set forth in claim 13, wherein the diaphragm is ruptured by the gas pressure of the second gas.

15. The process set forth in claim 13, wherein the diaphragm is ruptured by mechanical means.

16. A process for producing acetylene from hydrocarbons comprising the steps of introducing such hydrocarbons into an elongated reaction vessel, subjecting such hydrocarbons to the action of a shock wave and its associated rapid rise and fall of temperature and pressure sufficient to penetrate through said hydrocarbons to cause cracking of said hydrocarbons and transform a portion thereof into acetylene, and removing from the reaction vessel the reaction products of which said acetylene constitutes a part.

References Cited in the file of this patent

**UNITED STATES PATENTS**

- 2,475,282 Hasche ------------ July 5, 1949
- 2,690,960 Kistiakowsky et al. ------ Oct. 5, 1954
- 2,832,666 Hertzberg et al. -------- Apr. 29, 1958

**FOREIGN PATENTS**

- 737,555 Great Britain ---------- Sept. 28, 1955