Second Edition.]

PATENT SPECIFICATION

Application Date: Feb. 6, 1928. No. 3664/28. 310,999

Complete Left: Nov. 5, 1928.

Complete Accepted: May 6, 1929.

1189

PROVISIONAL SPECIFICATION.

Improvements in and Apparatus for the Manufacture and Production of Gaseous and Readily Volatile Olefines from Hydrogen and Oxides of Carbon.

I, JAMES YATE JOHNSON, a British Subject, of 47, Lincoln's Inn Fields, in the County of London, Gentleman, do hereby declare the nature of this invention (which has been communicated to me from abroad by I. G. Farbenindustrie Aktiengesellschaft, of Frankfort-on-Main, Germany, a Joint Stock Company organized under the Laws of Germany) to be as follows:—

The production of gaseous and readily volatile olefines, especially ethylene, from mixtures of oxides of carbon with hydrogen, at an elevated temperature and in the presence of catalysts, is attended, in practice, with considerable difficulties, inasmuch as the catalysts which at first furnish good yields, soon lose their efficiency, more particularly owing to the deposition of loose, flocculent carbon.

My foreign correspondents have now found that these difficulties are obviated and good regular yields of olefines, such as ethylene, obtained if care be taken to maintain the catalysts as far as possible at uniform temperature. According to this invention, this is accomplished by bringing the catalysts into conjunction with a flowing medium, in a manner facilitating heat conduction, the said medium being capable of supplying or removing heat. According to this manner of working the heat developed in the catalysts, by the reaction, is immediately carried away therefrom and injurious local superheating is avoided.

The catalysts may for example be disposed in a thin layer on a bedding of materials which readily conduct heat, such as copper, or an alloy of manganese and copper and like minerals, the opposite surface of which is in contact with flowing hot gases, hot liquids, or with saline or metallic melts and the like. Use may be made, for example, of copper tubes, manganese copper tubes, or copper blocks, provided with bores for the accommodation of the catalyst, and bores for receiving the flowing medium, or pieces of manganese copper provided with gills and bores, may also be used. The catalysts chiefly coming under consideration comprise finely divided or granular copper, silver, gold, iron and the like or mixtures thereof.

The present invention has the great advantage that any sudden rise in temperature is prevented. If the catalyst becomes too hot at any one place, it parts with its heat to the bedding, and the medium flowing through the latter absorbs such heat and carries it away.

I will describe two Examples of apparatus according to this invention which further explain the nature of the invention, reference being made to the accompanying drawing in which:—

Figure 1 shews in vertical section a high-pressure furnace with inserted manganese copper tube unit. Arriving from the superheater A, superheated steam enters the tube unit at B, flows straight up as far as C, and then flows downward through the several coiled tubes D on which is disposed a thin layer of a granular catalyst consisting of iron and uranium oxide. By means of the superheated steam, the furnace can be maintained at from 360 to 370 degrees Centigrade. A gas containing 2 per cent of carbon dioxide 23 per cent of carbon monoxide, 71 per cent of hydrogen and 4 per cent of nitrogen is passed through the furnace, from the top, under a pressure of about 120 atmospheres. The final gas contains uniformly 3 to 4 per cent of ethylene, propylene, butylene and amylene, with small amounts of ethane and propane. Water and a little oil are also formed. At the end of three weeks, no deposition of carbon can be detected on the catalyst.

In large furnaces it is advisable to arrange the hot steam conduit as a self contained circuit and to circulate the steam through the furnace and superheater by means of a blower. In this case, very little heat need be supplied to the superheater. The steam may also be passed upward through the coils, in which case the heat of reaction originating in the lower layers of catalyst will be carried

up by the steam and given off to the cold fresh gas through the upper coils D. The upper coils D may also be used solely for heating up, the layer of catalyst being omitted there in such case.

Figures 2 and 3 (being vertical sections at right angles to each other) shew a high-pressure furnace divided into four compartments one above the other, with gilled shelves of manganese copper A disposed between them. The catalyst B is placed between the gills. C are bores in the said shelves of manganese copper A, through which superheated steam from the superheater D is passed at a temperature of 300° Centigrade. A gaseous mixture of the kind specified in the preceding example is passed through the furnace by means of the openings E under a pressure of about 120 atmospheres. The temperature can be maintained regularly at about 360° Centigrade and the yield of ethylene and the like is the same as in Example I. There is no deposition of carbon on the catalyst. The several compartments can also be provided with their own separate steam supply and removal pipes, so that the temperature in each compartment can be controlled separately.

Dated this 6th day of February 1928.

JOHNSONS & WILLCOX,
47, Lincoln's Inn Fields, London.
W.C. 2,
Agents.

COMPLETE SPECIFICATION.

Improvements in and Apparatus for the Manufacture and Production of Gaseous and Readily Volatile Olefines from Hydrogen and Oxides of Carbon.

I, JAMES YATE JOHNSON, a British Subject, of 47, Lincoln's Inn Fields, in the County of London, Gentleman, do hereby declare the nature of this invention (which has been communicated to me from abroad by I. G. Farbenindustrie Aktiengesellschaft, of Frankfort-on-Main, Germany, a Joint Stock Company organized under the Laws of Germany) and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

The production of gaseous and readily volatile olefines, especially ethylene, from mixtures of oxides of carbon with hydrogen, at an elevated temperature and in the presence of catalysts, is attended, in practice, with considerable difficulties, inasmuch as the catalysts which at first furnish good yields, soon lose their efficiency, more particularly owing to the deposition of loose, flocculent carbon.

My foreign correspondents have now found that these difficulties are obviated and good regular yields of olefines, such as ethylene, obtained if care be taken to maintain the catalysts as far as possible at uniform temperature. According to this invention, this is accomplished by bringing the catalysts into conjunction with a flowing medium, in a manner facilitating heat conduction, the said medium being capable of removing surplus heat and not being in contact with the reacting gases. According to this manner of working the heat developed in the catalysts, by the reaction, is immediately carried away therefrom and injurious local superheating is avoided.

The catalysts may for example be disposed in a thin layer on a bedding of a material which readily conducts heat, such as copper, or an alloy of manganese and copper and like materials, the opposite surface of which is in contact with flowing gases, liquids, or with saline or metallic melts and the like the temperature of which is lower than the reaction temperature. Use may be made, for example, of copper tubes, manganese-copper tubes, or copper blocks, provided with bores for the accommodation of the catalyst, and bores for receiving the flowing medium, or peices of manganese copper provided with gills and bores, may also be used. The catalysts chiefly coming under consideration comprise finely divided or granular metals, for example, copper, silver, gold, iron and the like or mixtures thereof.

The reaction is usually carried out at between about 200° and 500° Centigrade and preferably between 350° and 400° Centigrade, and as a rule under elevated pressure, say at about 100 atmospheres, but higher pressures, for example 150, 200 or even more atmospheres or lower pressures, for example 50 or 20 atmospheres may also be used. Also atmospheric pressure comes into question.

The aforesaid flowing media may also be employed for preheating the gaseous mixtures to be subjected to the reaction.

The present invention has the great

advantage that any sudden rise in temperature is prevented. If the catalyst becomes too hot at any one place, it parts with its heat to the bedding, and the medium flowing through the latter absorbs such heat and carries it away.

I will describe two Examples of apparatus according to this invention which further explain how the said invention may be carried into practical effect, reference being made to the drawings accompanying the Provisional specification in which :—

Figure 1 shews in vertical section a high-pressure reaction furnace with inserted manganese-copper tube unit. Arriving from the superheater A, superheated steam enters the tube unit at B, flows straight up as far as C, and then flows downward through the several coiled tubes D on which is disposed a thin layer of a granular catalyst consisting of iron and uranium oxide. By means of the superheated steam, the reaction furnace can be maintained at from 360° to 370° Centigrade. A gas containing 2 per cent of carbon dioxide 23 per cent of carbon monoxide, 71 per cent of hydrogen and 4 per cent. of nitrogen is passed through the furnace, from the top, under a pressure of about 120 atmospheres. The final gas contains uniformly 3 to 4 per cent. of ethylene, propylene, butylene and amylene, with small amounts of ethane and propane. Water and a little oil are also formed. At the end of three weeks, no deposition of carbon can be detected on the catalyst.

In large furnaces it is advisable to arrange the hot steam conduit as a self-contained circuit and to circulate the steam through the reaction vessel and superheater by means of a blower. In this case, very little heat need be supplied to the superheater. The steam may also be passed upward through the coils, in which case the heat reaction originating in the lower layers of catalyst will be carried up by the steam and given off to the cold fresh gas through the upper coils D. The upper coils D may also be used solely for heating up the mixture to be subjected to the reaction, the layer of catalyst being omitted there in such case.

Figures 2 and 3 (being vertical sections at right angles to each other) shew a high-pressure reaction vessel divided into four compartments one above the other, with gilled shelves of manganese copper A disposed between them. The catalyst B is placed between the gills. C are bores in the said shelves of manganese copper A, through which superheated steam from the superheater D is passed at a temperature of 360° Centigrade. A gaseous mixture of the kind specified in the preceding example is passed through the reaction vessel by means of the openings E under a pressure of about 120 atmospheres. The temperature can be maintained regularly at about 360° Centigrade and the yeild of ethylene and the like is the same as described with reference to Figure 1. There is no deposition of carbon on the catalyst. The several compartments can also be provided with their own separate steam supply and removal pipes, so that the temperature in each compartment can be controlled separately.

Having now particularly described and ascertained the nature of my said invention and in what manner the same is to be performed, I declare that what I claim is :—

1. A process for the manufacture and production of gaseous and readily volatile olefines from mixtures of oxides of carbon and hydrogen at an elevated temperature and in the presence of catalysts, which consists in bringing the catalysts into conjunction with flowing medium in a manner facilitating heat conduction, the said medium being capable of removing surplus heat and not being in contact with the reacting gases.

2. The specific method of carrying out the process according to claim 1, which consists in disposing the catalyst in thin layers on a bedding of a material which readily conducts heat, the opposite surface of which is in cantact with the medium capable of removing surplus heat.

3. The modification of the processes claimed in claims 1 and 2, which consists in employing the said media capable of removing surplus heat also for preheating the gaseous mixtures to be subjected to the reaction.

4. The process for the manufacture and production of gaseous and readily volatile olefines substantially as hereinbefore described and illustrated with reference to the apparatus described in the accompanying drawings.

5. Gaseous and readily volatile olefines when obtained according to the processes of the preceding claiming clauses.

6. Apparatus for the production of gaseous and readily volatile olefines substantially as hereinbefore described and illustrated in the accompanying drawing.

Dated this 5th day of November, 1928.

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Agents.

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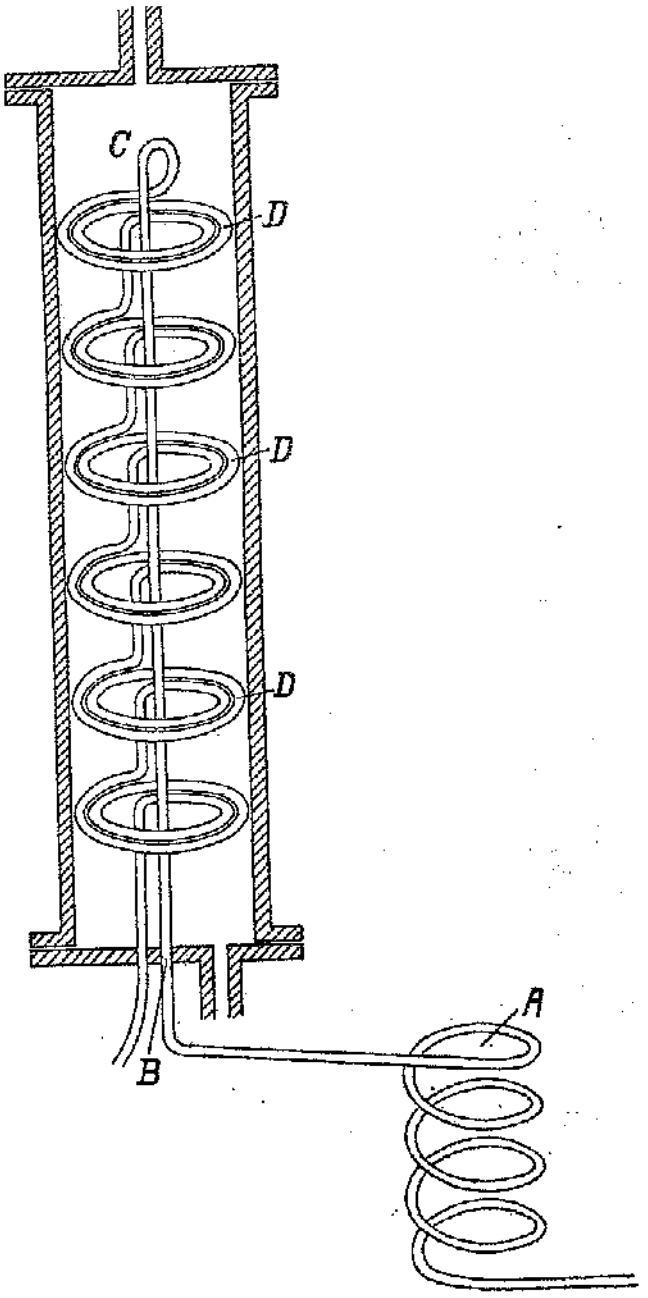
2nd Edition

Fig. 1



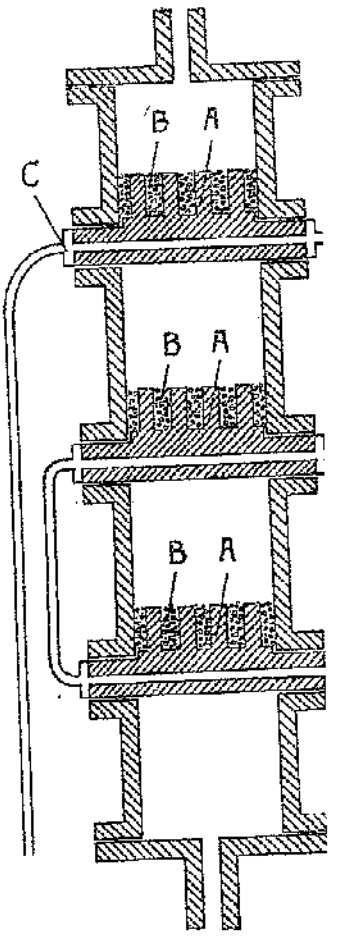


Fig.2

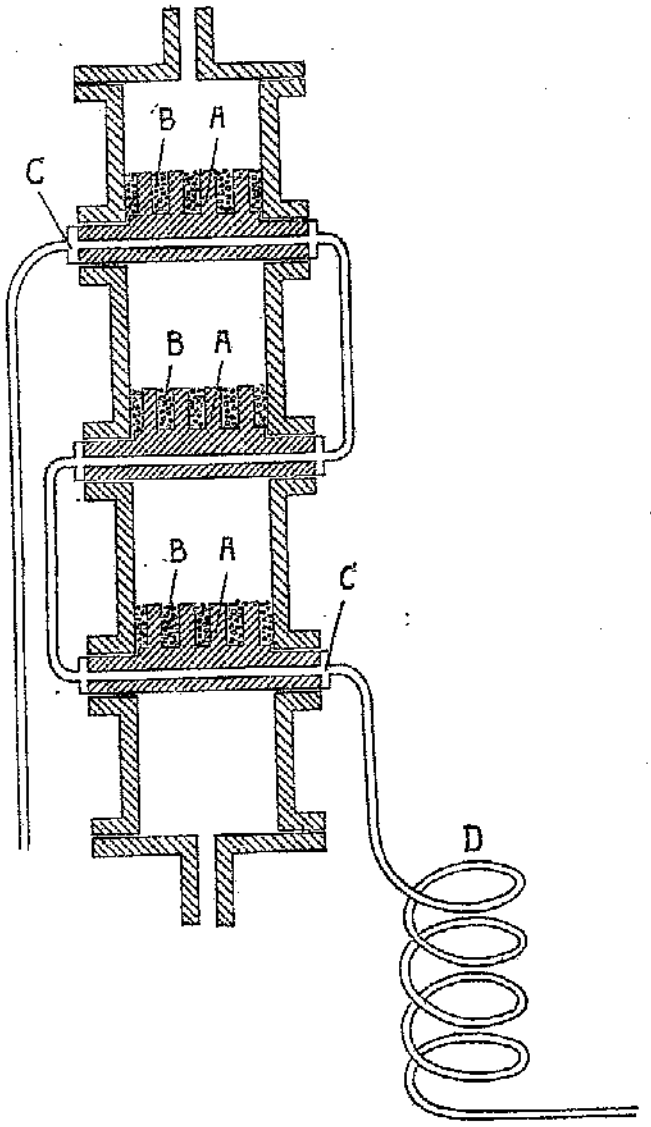
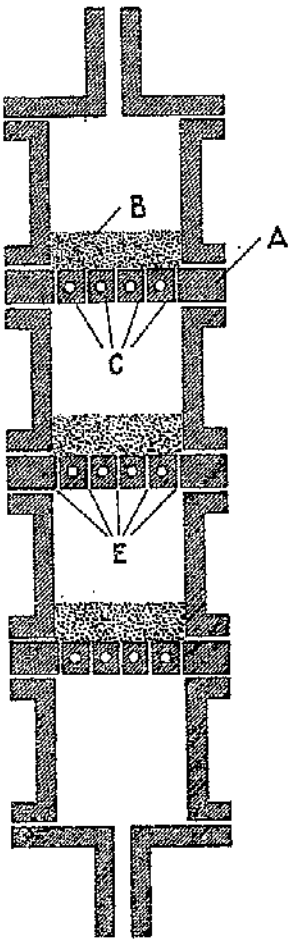


Fig.3.



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810,999 PROVISIONAL SPECIFICATION

2nd Edition

[This Drawing is a reproduction of the Original on a reduced scale]

Fig.1

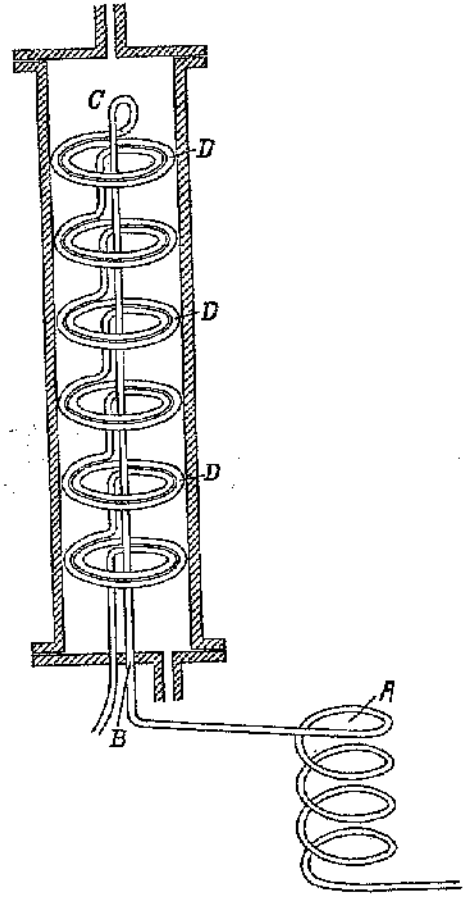


Fig.2

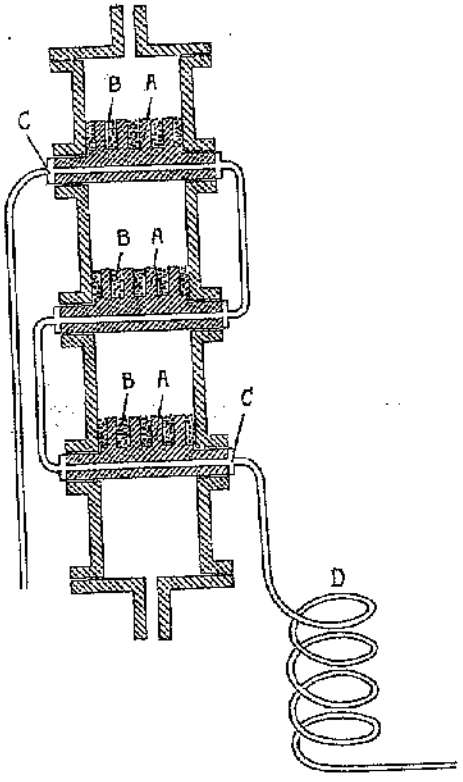
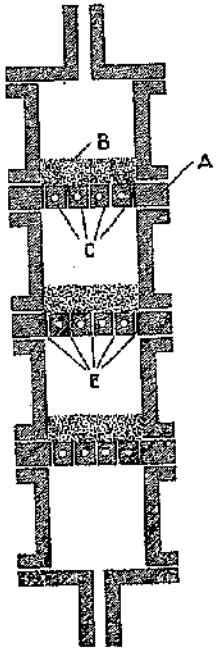


Fig.3.



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