[Second Edition.]

PATENT SPECIFICATION

Application Date: April 8, 1929. No. 10,805/29. **336,635**

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PROVISIONAL SPECIFICATION.

Improvements in the Manufacture and Production of Hydrogen and Gases Containing Hydrogen.

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:—

My foreign correspondents have found that the conversion of gaseous hydrocarbons such as methane, or gas mixtures containing the same into hydrogen and carbon, or in the presence of water vapour or oxygen or both into mixtures of hydrogen and carbon monoxide by heating at elevated temperatures is effected in a very simple and economical manner when the same is undertaken in one operation with the production of gas by the carbonisation of coal or by the gasification of coal or coke or other carbonaceous material.

It is preferable to operate by passing the gas mixture containing hydrocarbons, which is to be converted, either as such, or together with water vapour or air or both through the hot layer of coal which is degasified to a more or less great extent and of coke and which in any case has a temperature of at least from about 800° to 1000° Centigrade. In this manner the degasification of the layer of coal and coke takes its course and at the same time the hydrocarbons introduced are decomposed with the formation of hydrogen, and in cases when sufficient amounts of water vapour or oxygen are present with the formation of hydrogen and the oxides of carbon. In some cases the process may even be carried out when the coal is already degasified. The gas mixture leaving the coke furnace may be partially or wholly freed from constituents other than hydrogen in order to obtain hydrogen or gas mixtures rich in hydrogen, or it may be employed without further treatment.

For example, by leading methane itself or gases containing methane or other hydrocarbons together with water vapour from above or below through the column of glowing coke in a water gas producer, the methane or other hydrocarbons are converted into hydrogen and carbon monoxide and small amounts of carbon dioxide, and a gas mixture rich in hydrogen, which consists of these gases together with the water gas, which is produced at the same time is obtained, and this may be worked up into pure hydrogen by the removal of the oxides of carbon and the other substances mixed therewith, or it may be employed without further treatment.

The gases containing hydrocarbons to be converted may be derived from any source, for example they may be of natural origin. Thus, for example, the manufacture of hydrogen by the decomposition of natural gas in the manner hereinbefore described may be carried out very advantageously in coke ovens. It has also been found to be advantageous to work up the waste gases from the synthesis of ammonia in this manner. In the synthesis of ammonia from its elements while leading the reaction gas mixture in a cycle, the gas mixture gradually becomes so enriched with methane, that a part of the mixture must be continuously or periodically removed from circulation in order to reduce the methane content and in this manner considerable amounts of the gases for the synthesis are lost to the process. By means of the process according to the present invention it is now possible to avoid this loss because the gas containing methane, which is withdrawn from circulation, may be passed through a water gas producer in the manner already described and may be returned together with the water gas produced at the same time to the synthesis of ammonia after the carbon monoxide contained in the mixture has been converted with steam in the manner already known into hydrogen and carbon dioxide and the latter has been removed, and if necessary after replenishing the nitrogen or hydrogen content. Moreover the waste gases from the destructive hydrogenation of coal, and similar hydrogenation processes, which are frequently rich in gaseous hydrocarbons such as methane, may be converted in the manner

hereinbefore described into valuable gases rich in hydrogen.

The conversion of the gases containing hydrocarbons into gas mixtures containing hydrogen or hydrogen and carbon monoxide, by the process in accordance with the present invention, may be carried out in a similar manner in gas producers other than water gas producers, for example it may be carried out together with the production of producer gas. In this case a gas mixture containing hydrogen, which accordingly has a different composition, is obtained.

The following Examples will further illustrate the nature of this invention but the invention is not restricted to these Examples.

EXAMPLE 1.

Through the glowing column of coke in a water gas producer, in which by working according to the usual method about 4000 cubic metres per hour of a gas having the following composition are obtained: 4.0 per cent of CO_2, 41.0 per cent of CO, 46.0 per cent of H_2, 0.2 per cent of CH_4 and 8.8 per cent of N_2, are passed together with the steam about 1000 cubic metres per hour of waste gas from the ammonia synthesis having the composition: 10 per cent of CH_4, 70 per cent of H_2 and 20 per cent of N_2. In this manner about 5300 cubic metres per hour of a gas mixture having the following composition are obtained. 4.0 per cent of CO_2, 32.0 per cent of CO, 53.5 per cent of H_2, 0.2 per cent of CH_4, and 10.3 per cent of N_2.

EXAMPLE 2.

1000 cubic metres of waste gas from the synthesis of ammonia consisting of 10 per cent of methane, 70 per cent of hydrogen and 20 per cent of nitrogen are passed per hour together with 200 kilograms of steam through the glowing column of a coke chamber. About 1470 cubic metres of a gas mixture having the approximate composition: 3.8 per cent of carbon dioxide, 8.0 per cent of carbon monoxide, 73.5 per cent of hydrogen, 0.2 per cent of methane and 14.5 per cent of nitrogen are obtained.

Dated this 8th day of April, 1929.

J. Y. & G. W. JOHNSON,
47, Lincoln's Inn Fields, London, W.C. 2,
Agents.

COMPLETE SPECIFICATION.

Improvements in the Manufacture and Production of Hydrogen and Gases Containing Hydrogen.

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:—

My foreign correspondents have found that the conversion into hydrogen and carbon monoxide, of gaseous hydrocarbons such as methane, or gas mixtures containing the same, other than coke oven gases or other gases containing liquid hydrocarbons, in admixture with water vapour and if desired with oxygen, by heating at elevated temperatures is effected in a very simple and economical manner when the said conversion is carried out by introducing the initial materials into the charge in a water gas producer in which coke or other carbonaceous material is undergoing gasification the said gasification being carried out without an addition of substantial amounts of carbon dioxide, the conversion of the said gaseous hydrocarbons proceeding simultaneously with the said gasification.

It is preferable to operate by passing into the producer the gas mixture containing hydrocarbons, which is to be converted, together with water vapour and if desired with air through the hot layer of coke which has a temperature of at least from about 800° to 1000° Centigrade. In this manner the gasification of the layer of coke takes its course and at the same time the hydrocarbons and steam introduced are decomposed with the formation of hydrogen, and the oxides of carbon. The gas mixture leaving the producer may be partially or wholly freed from constituents other than hydrogen in order to obtain hydrogen or gas mixtures rich in hydrogen, or it may be employed without further treatment.

The gases containing hydrocarbons to be converted may be derived from any source, for example they may be of natural origin. It has also been found to be advantageous to work up the waste gases containing hydrocarbons from industrial processes, such as the synthesis

of ammonia in this manner. In the synthesis of ammonia from its elements while leading the reaction gas mixture in a cycle, the gas mixture gradually becomes so enriched with methane, that a part of the mixture must be continuously or periodically removed from circulation in order to reduce the methane content and in this manner considerable amounts of the gases for the synthesis are lost to the process. By means of the process according to the present invention, it is now possible to avoid this loss because the gas containing methane, which is withdrawn from circulation, may be passed through a water gas producer in the manner already described and may be returned together with the water gas produced at the same time to the synthesis of ammonia after the carbon monoxide contained in the mixture has been converted with steam in the manner already known into hydrogen and carbon dioxide and the latter has been removed, and if necessary after replenishing the nitrogen or hydrogen content. Moreover the waste gases from the destructive hydrogenation of coal, and similar hydrogenation processes or gases derived from cracking processes, which are frequently rich in gaseous hydrocarbons such as methane, may be converted in the manner hereinbefore described into valuable gases rich in hydrogen.

The process in accordance with the present invention is attended by the particular advantage that for the conversion of gaseous hydrocarbons into gases containing hydrogen, as for example for the conversion of waste gases containing methane, no special apparatus is required and the said conversion can be carried out in a water gas producer already existing. A further advantage of the present process is that it is continuous and can be carried out without interruption of the working of the said gas producers.

The following Example will further illustrate how the said invention may be carried out in practice but the invention is not restricted to this Example.

EXAMPLE.

Through the glowing column of coke in a water gas producer, in which by working according to the usual method about 4000 cubic metres per hour of a gas having the following composition are obtained: 4.0 per cent of CO_2, 41.0 per cent of CO, 46.0 per cent of H_2, 0.2 per cent of CH_4 and 8.8 per cent of N_2 steam is passed during the cold blowing period together with about 1000 cubic metres per hour of waste gas from the ammonia synthesis having the composition: 10 per cent of CH_4, 70 per cent of H_2 and 20 per cent of N_2. In this manner about 5300 cubic metres per hour of a gas mixture having the following composition are obtained from the said mixture of steam and waste gas from the ammonia synthesis: 4.0 per cent of CO_2, 32.0 per cent of CO, 53.5 per cent of H_2, 0.2 per cent of CH_4 and 10.3 per cent of N_2.

I am aware of Specification No. 279,072 and do not claim anything described or claimed therein.

It has been proposed to use solid fuels such as wood, charcoal, coal or coke as incandescent masses for facilitating the decomposition of methane and oxygen into hydrogen and carbon monoxide.

I am also aware of the proposition to produce an enriched or carburetted water gas by introducing vaporized liquid hydrocarbons, such as liquid fuels or crude or waste oils into a generator or producer, together with steam. The present invention exclusively relates to the conversion of gaseous hydrocarbons.

I am also aware that it has been suggested to carbonize coal by means of the heat contained in the gases emanating from water gas producers and employing an auxiliary gas such as oil gas or other gases containing liquid hydrocarbons during the blowing periods of the water gas producer as a means of carrying the heat required for carbonization directly to the material in the carbonizing chamber while, during the gas making periods, the water is employed in the same manner, both gases being heated to a suitable temperature before entering the distilling retort.

I am aware of specifications Nos. 286,291, 188,494 and 17,053/99 and I make no claim to anything described or claimed therein.

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 production of hydrogen and carbon monoxide by conversion of gaseous hydrocarbons, such as methane or gas mixtures containing the same, which consists in introducing the said gaseous hydrocarbons, other than coke oven gases or other gases containing liquid hydrocarbons with an addition of water vapour and if desired with air or oxygen or mixtures containing these gases, into the charge in a water gas producer in which coke or other carbonaceous material is undergoing gasification, the said gasification being carried out without an addition of substantial amounts of carbon dioxide, the conversion of the gaseous hydrocarbons proceeding simultaneously with

the said gasification.

2. A modification of the process claimed in Claim 1 which comprises introducing the waste gases containing hydrocarbons from the synthesis of ammonia, into the charge in a water gas producer in which coke or other carbonaceous material is undergoing gasification.

3. The process for the conversion into hydrogen and carbon monoxide of gaseous hydrocarbons, such as methane, or gas mixtures containing the same in admixture with water vapour and if desired with oxygen by heating at elevated temperatures, substantially as described in the foregoing Examples.

4. In the process as claimed in Claims 1 and 2, the employment of waste gases containing hydrocarbons from industrial processes, such as the destructive hydrogenation of carbonaceous materials or the synthesis of ammonia as the initial materials.

5. Hydrogen and carbon monoxide, when obtained according to the preceding claiming clauses.

Dated this 18th day of December, 1929.

J. Y. & G. W. JOHNSON,
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Agents.

Reference has been directed, in pursuance of Section 7, Sub-section 4, of the Patents and Designs Acts, 1907 to 1928, to Specification No. 101,152.

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