[Second Edition].

PATENT SPECIFICATION

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PROVISIONAL SPECIFICATION.

Improvements in the Manufacture and Production of Benzene and its Homologues from Mixtures of Oxides of Carbon and 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 mixtures of oxides of carbon with hydrogen such as water gas and the like may be converted, with a good yield, into benzene and its homologues if the said gaseous mixtures be first converted, by the aid of catalysts, into gaseous heavy hydrocarbons and gaseous homologues of methane, such as ethane and the like, and the resulting gaseous hydrocarbons be converted in any known manner with or without the application of pressure and in the presence or absence of catalysts, into benzene and its homologues.

Iron preferably mixed with noble metals and difficulty reducible oxides, is to be used as a contact mass for the first stage of the process, and the resulting gaseous mixtures containing gaseous hydrocarbons, may be further treated as such, preferably after being freed from any carbon dioxide present, or the gaseous hydrocarbons may be recovered by any known or suitable method, such as with the aid of active charcoal or by strong cooling, and subjected alone to the further treatment.

The following example will further illustrate the nature of the said invention which however is not limited thereto.

EXAMPLE.

Water gas, under a pressure of about 50 atmospheres, is passed at 370° Centigrade over an iron-silver contact mass at a speed of 40 litres per hour calculated on atmospheric pressure over each litre of the contact mass. A gas is formed which contains 61 per cent. of CO_2, 13 per cent. of C_nH_{2n}, 1.6 per cent. of CO, 2 per cent. of H_2, 10 per cent. of C_2H_6, 4 per cent. of CH_4 and 8.4 per cent. of N_2. This gas is cooled, its sensible heat being used for preheating the initial gases and is then passed quickly, at 30 atmospheres pressure, through a washing tower, fed with water of about 30° Centrigrade, which dissolves out about three-fourths of the carbon dioxide present. The gas is then expanded, and the remaining carbon dioxide is removed with ammonia water. The residual gas contains C_nH_{2n}, 32 per cent.; CO, 4 per cent.; H_2, 5 per cent.; C_2H_6, 25 per cent.; CH_4, 10 per cent.; N_2, 24 per cent. It is passed slowly at about 800° Centigrade, through a copper tube filled with copper turnings free from iron and is thereby converted almost quantitatively, and without deposition of carbon, into a liquid containing about 80 per cent. of benzene.

Dated this 13th day of July, 1927.

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

COMPLETE SPECIFICATION.

Improvements in the Manufacture and Production of Benzene and its Homologues from Mixtures of Oxides of Carbon and 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 particu-

larly described and ascertained in and by the following statement:—

My foreign correspondents have found that mixtures of oxides if carbon with hydrogen such as water gas and the like may be converted, with a good yield, into benzene and its homologues if the said gaseous mixtures be first converted, by the aid of catalysts, into gaseous heavy hydrocarbons and gaseous homologues of methane, such as ethane and the like, and the resulting gaseous hydrocarbons be converted in a second stage into benzene and its homologues.

The first stage of the process is a catalytic treatment in which the initial gases are passed at temperatures above 300° Centigrade and preferably between about 350° and 370° Centigrade over catalysts comprising iron. Particularly suitable catalysts are mixtures of iron with noble metals, or difficultly reducible oxides of metals, or both. The process at this stage may be carried out at atmospheric pressure, but it is preferable to work at an elevated pressure, for example, 10, 20 or 50 atmospheres. Though even higher pressures may be employed, it is generally not advantageous to increase the pressure to above 100 atmospheres. The gases resulting from the said catalytic treatment contain ample amounts of gaseous olefines and gaseous homologues of methane, such as ethane. A method for carrying out this stage of the process has been described and claimed in Specification No. 293,572 of even date herewith.

The said gases are then subjected to the second stage-treatment which consists in exposing them to the action of temperatures between about 600° and 800° Centigrade. The gases are subjected to this treatment preferably after removing any carbon dioxide present therein, or the gaseous olefines and gaseous homologues of methane may be separated from the gas mixture resulting from the first stage in any suitable manner and subjected alone to the second stage treatment. The said second stage treatment is preferably carried out in the absence of materials giving rise to a deposition of carbon, such as iron. Most suitably it is carried out with the aid of catalysts, such for example as copper, and also the material of the apparatus may act as catalyst. The apparatus may be constructed, for example, from copper, or from copper or iron lined or coated with tin, zinc, or chromium. The second stage of this process may be carried out at atmospheric pressure as well as at higher pressures. A method for the operation of this stage of the process is described and claimed in Specification No. 258,608.

The following example will further illustrate how the said invention may be carried out in practice but the invention is not limited thereto.

EXAMPLE.

Water gas, under a pressure of about 50 atmospheres, is passed at 370° Centigrade over an iron-silver catalyst, prepared by melting a mixture of fine iron powder with 10 per cent. its weight of silver nitrate in a current of oxygen and reducing the product in a current of hydrogen at about 450° to 500° Centigrade at a speed of 40 litres calculated on atmospheric pressure per hour and over each litre of the contact mass. A gas is formed which contains 61 per cent. of CO_2, 13 per cent, of C_nH_{2n}, 1.6 per cent. of CO, 2 per cent. of H_2, 10 per cent. of C_2H_6, 4 per cent. of CH_4 and 8.4 per cent. of N_2. This gas is cooled its sensible heat being used for repeating the initial gases and is then passed quickly, at 30 atmospheres pressure, through a washing tower, fed with water of about 30° Centigrade which dissolves out about three-fourths of the carbon dioxide present. The gas is then expanded, and the remaining carbon dioxide is removed with ammonia water. The residual gas contains C_nH_{2n}, 32 per cent.; CO, 4 per cent.; H_2, 5 per cent.; C_2H_6, 25 per cent.; CH_4, 10 per cent.; N_2, 24 per cent. It is passed slowly, at a rate of about 60 litres per hour for each litre capacity of the reaction vessel, at about 800° Centigrade through a copper tube filled with copper turnings free from iron and is thereby converted almost quantitatively, and without deposition of carbon, into a liquid containing about 80 per cent. of benzene.

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 benzene and its homologues which consists in passing in a first stage a mixture of an oxide of carbon and hydrogen, preferably at elevated pressure, at a temperature above 300° Centigrade over a catalyst comprising iron and heating in a second stage the resulting gaseous olefines and gaseous homologues of methane under atmospheric or elevated pressure to temperatures, between about 600° and 800° Centigrade in the presence or absence of a catalyst.

2. A modification of the process in accordance with Claim 1 which consists in employing in the first stage a catalyst comprising iron and a noble metal, or a difficultly reducible oxide of a metal, or a mixture thereof.

3. A modification of the process in accordance with the preceding claiming clauses which consists in carrying out the second stage in the absence of materials giving rise to a decomposition of carbon, particularly of iron.

4. The process for the production of benzene and its homologues substantially as described in the foregoing example.

5. Benzene and its homologues when prepared in accordance with the preceding claiming clauses.

Dated this 13th day of April, 1928.

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