[Second Edition.]

PATENT SPECIFICATION

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COMPLETE SPECIFICATION.

Process of Obtaining Aromatic Hydrocarbons.

I, ALEXANDER SYDNEY RAMAGE, of 7644, Woodward Avenue, Detroit, Michigan, United States of America, a citizen of the United States of America, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

This invention relates to the production of aromatic hydrocarbons.

Numerous proposals have been made in connection with the treatment of mixtures of water gas or the component gases thereof in association with open chain olefinic, acetylenic and other unsaturated hydrocarbons by contacting the mixture at atmospheric or super atmospheric pressure and at raised temperatures, for instance, a temperature of 500° C. with a catalyst, for example, iron which may be in pulverulent or other suitable form the object being to obtain heavier, aromatic or other hydrocarbons from lighter hydrocarbons.

In a process for the manufacture of liquid fuels having the general properties of petrols and benzols, gases rich in hydrocarbons, particularly of the acetylene series, are added to water gas issuing from a producer, and the complex gas mixture thus obtained is passed over a hydrogenating catalyst, then over a dehydrogenating catalyst, and finally over a third catalyst consisting, for instance, in pumice stone charged with iron in order to effect polymerisation of the acetylene hydrocarbons.

The process according to the present invention comprises contacting a mixture containing unsaturated hydrocarbons and a gas consisting essentially of hydrogen and carbon monoxide, for instance, water gas with metallic iron at a temperature of from 1000° F. to about 1500° F. at substantially atmospheric pressure.

Thus, the mixture may be passed through a series of tubes containing metallic iron in the form, for example, of turnings or in a finely divided reduced state.

Furthermore, the proportion of unsaturated hydrocarbons employed relatively to the hydrogen and carbon monoxide is such that the iron is maintained in an active condition and to this end a proportion of unsaturated hydrocarbon gas in excess of that required for the formation of aromatic hydrocarbons is maintained in the gaseous mixture.

Preferably, in accordance with the invention, the unsaturated hydrocarbons employed are hydrocarbons of the olefine series which may be derived from any suitable source, for instance, by dehydrogenating light weather products and casing head gasoline vapours over ferric oxide or cupric oxide.

The mixture of hydrogen and carbon monoxide may be obtained, for instance, from a standard water gas producer, or the mixture may be prepared from hydrogen and carbon monoxide obtained from any other source.

The reactions taking place in the process are complicated and not fully known but the issuing gases contain large quantities of the hydrocarbons of the aromatic series including benzol, toluol, xylol, etc., which can be condensed and washed out of the gases by any of the standard methods. The gas from the wash oil scrubbers after the extraction of the aromatic hydrocarbons is of very high heat value running generally over 1000 B.T.U. and can be utilised for mixing with gases of low heat value to carburet them. The reactions may be as follows:—

The metallic iron presumably acts as a catalyst in transferring the oxygen of the carbon monoxide to the unsaturated hydrocarbons which, as stated above, must be in such quantity as to keep the iron in the active condition.

The reduced metallic iron in the tubes ordinarily requires no attention, but may, if for any reason it becomes oxidised or contaminated with impurities, be renewed by suitable treatment with reducing gases or replaced by fresh material.

The unsaturated hydrocarbon gases and vapours subjected to this treatment should be nearly or quite free from paraffin hydrocarbons, as, if these are present in any considerable amount, cracking results and the accompanying deposition of

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carbon tends to plug up the exit tubes or otherwise to interfere with the operation.

In order to ensure the substantial absence of any paraffin hydrocarbons, it may even be desirable to subject the gases to a preliminary treatment by passing them at a suitable temperature over ferric oxide or cupric oxide by which such paraffin hydrocarbons will be converted into unsaturated hydrocarbon gases.

The accompanying diagrammatic drawing illustrates a plant for carrying out the process when employing hydrocarbons of the olefine series.

Gases 1 of the olefine series mixed with water gas 2 or its components are passed through a "converter" 3 which comprises a series of tubes containing metallic iron, heated by burner 4 to the temperatures above described. Products of combustion leave the converter furnace by way of the flue 16. The vapours from the converter are passed through a condenser 5 and separator 6 for the removal of any condensable aromatics. The cooled gas from the separator 6 is delivered by pump 7 to the tower 8 where it is scrubbed by contact with a suitable wash-oil introduced at the top of the tower by the pump, 9. The absorbent wash-oil is then delivered by pump 10, preferably through a heat exchanger 11, to a still 12 in which the aromatics and other volatile ingredients are driven off and condensed by condenser 13. The residual oil from the still 12 passes through the heat-exchanger 11 to the top of the scrubbing tower 8, from which the scrubbed gas 14 goes to a holder or place of use.

The condensate from the wash-oil is preferably mixed in receptacle 15 with the condensate from the separator 6. The mixed condensate may be used for blending with gasoline to produce a high-compression motor fuel or it can be refined in the well-known manner for the production of pure benzol, toluol, xylol etc.

I prefer to use 3 inch tubes 10 feet long set in manifolds, the manifolds set preferably in groups of three. The gases enter the lower manifold then pass through the middle manifolds and finally through the upper manifold so that they pass through 30 feet of tube filled with metallic iron. The manifolds are set in the usual way in a combustion chamber, the heating gases preferably passing downward over the upper manifold first and issuing below the lowest manifold. In this way the upper manifold naturally is hotter than the lower. A suitable mixture of gases per tube hour is 600 cu. ft. of water gas and 400 cu. ft. of olefinic hydrocarbon gases of the average composition represented by the formula C_4H_8. With this amount per hour per tube the total length of the tubes preferably is, as stated above, about 30 feet and the temperature in the lowest or entry manifold is in the neighbourhood of 1000° F., for instance 1100° F. and in the upper or issuing manifold about 1200° F. If the temperatures are increased to 1400° F. and 1500° F. respectively, the amount of gas passed through each tube must be increased to about 1500 cu. ft. per hour. If the olefinic gases are of lower molecular weight, the proportion thereof to water gas in the mixture must be increased accordingly bearing in mind that the regulating feature is that the iron must be maintained always in the active condition.

The gas issuing from the manifolds is characterised by the substantial absence of CO_2 usually less than 1 per cent. which fact seems to show that iron which may have been oxidised is not reduced by CO, but the oxygen is transferred to the hydrocarbon present in the gas mixture.

In order to keep the iron in the metallic and active state an excess of gaseous olefine hydrocarbons equivalent preferably to at least 5 per cent. of the resultant hydrocarbons issuing from the scrubbing tower must be maintained in the mixture.

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 aromatic hydrocarbons which comprises contacting a mixture containing unsaturated hydrocarbons and a gas consisting essentially of hydrogen and carbon monoxide, for instance, water gas with metallic iron at a temperature of from 1000° F. to about 1500° F. at substantially atmospheric pressure.

2. A process as claimed in Claim 1 in which the mixture is passed through a series of tubes containing metallic iron in the form, for example, of turnings or in a finely divided reduced state.

3. A process as claimed in either of the preceding claims in which the proportion of unsaturated hydrocarbons employed relatively to the hydrogen and carbon monoxide is such that the iron is maintained in an active condition.

4. A process as claimed in any of the preceding claims in which the unsaturated hydrocarbons employed are hydrocarbons of the olefine series.

5. The process for the production of aromatic hydrocarbons substantially as hereinbefore described.

Dated this 3rd day of September, 1929.

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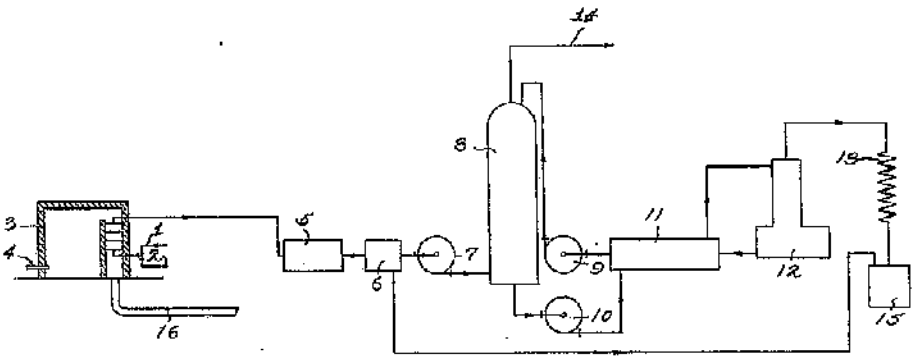
2nd Edition



[This Drawing is a full-size reproduction of the Original.]

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