SPECIFICATION



Application Date: Dec. 30, 1927. No. 36,287 127.

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Complete Accepted: April 2, 1929.

PROVISIONAL SPECIFICATION.

309,002

Process for the Synthesis of Liquid Hydrocarbons.

I Harold Edwin Potts, M.Sc., Chartered Patent Agent, of 12, Church Street, Liverpool, in the County of Lancaster, Subject of the King of Great Britain, do hereby declare the nature of this invention which has been communicated to me by Hermann Plauson, a Citizen of Esthonia, of Hagedornstrasse 51, Hamburg 37, Germany, to be as 10 follows :-

This invention relates to a process for the synthesis of liquid hydrocarbons of various boiling points from gases such as water gas, and/or gaseous hydrocarbons, 15 such as coke oven gas, natural gas or gases

produced by cracking heavy oils.

According to the present invention, mixtures of inflammable gaseous carbon compounds are converted into liquid hydro-carbons by subjecting the compounds to the action of rapidly moving electrons (βrays) and for to the reaction of X-rays.

An important feature of the invention consists in subjecting the reacting gases or vapours to the action of intense electric and for electro magnetic fields while under

the influence of the rays.

Further features of the invention consist in subjecting the activated gases during or especially after irradiation to the action of high pressure and/or temperatures in the latter case a "ripening" or after reaction occurs. The invention after reaction occurs. The invention further includes the introduction of suit-35 able catalysts into the reaction chamber. A gas mixture may be irradiated or one or more of the individual gases may be irradiated just prior to admixture, which is preferably followed by compression.

Another important feature of the inven-

tion is the use of mixtures of saturated and unsaturated hydrocarbons as the initial substances in the synthesis of

liquid hydrocarbons.

The invention also consists in the process of subjecting gaseous carbon compounds to the joint action of β -rays and X-rays, whereby liquid hydrocarbons are produced.

The β -rays may be produced by apparatus such as a Coolidge electron tube or that described in my co-pending application No. 14,539/27, or by apparatus constructed on similar principles. In this apparatus the electrons are emitted in large quantities from a surface coated with the elements or oxides of the elements of the rare earth series, heated electrically or otherwise, and enclosed in a highly evacuated chamber. A window of thin metal foil through which the rays can poss freely is provided in the chamber opposite the emitting surface and a reaction chamber is arranged externally to the window.

The β -rays must have sufficiently high velocities and concentrations. A velocity of between 200,000 and 200,000 kilometres per second is desirable and in all cases the per second is desirable and in all cases the velocity preferably exceeds one half the velocity of light. The voltage applied to the β -ray tube to accelerate the rays to this velocity varies between 50,000 and 500,000 volts. For most practical purposes a potential of 250—360,000 volts is sufficient. sufficient.

The admixture of X-rays may be produced from the action of the β -rays on such substances as glass, quartz, felspar, arranged above the metal window. The action of the β -rays on such substances produces X-radiation, the ionisation caused by the X-rays assisting the action

of the S-rays on the gases or vapours.

A main anode is arranged opposite to the metal window, and many form one wall of the reaction chamber. This anode is preferably maintained at a high positive potential, and the reacting substances pass between the anode and the metal window, so that they are simultaneously subjected to the action of β -rays, X-rays, and an intense electric field. An alternating voltage may however be applied to the anode.

A plurality of β -ray tubes may act in parallel on a single reaction chamber, and the gases may be introduced in the direction of, transversely, or in counter flow to the β -rays.

The power of β -rays to promote chemical reactions may be enhanced by causing the β-rays to rotate so that they traverse a spiral or helical path. Such a motion may be caused by allowing the rays to 105 traverse a rotating magnetic field. A

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rotating magnetic field may be produced by passing three phase alternating current through a system of three coils such as are used to produce a rotating magnetic field in alternating current motors. The combined effect of the rotating magnetic field and the velocity of the electrons forming the rays causes the rays to traverse a spiral or helical path. -

Catalysts may be used to accelerate the reactions. Suitable catalysts are those known in the heat-pressure synthesis of liquid hydrocarbons from water gas.

the present invention it is possible to increase the yield by circulating the reacting substances under increased pressures and/or temperatures e.g. to synthesise liquid hydrocarbons from water gas alone, a pressure of 200—1000 atmospheres is desirable; with a mixture of water gas and ethylene or acetylene, the reaction proceeds at ordinary pressures with a 20-30% yield and with increased pressures of 50-200 atmospheres approaches the theoretical yield. If desired, a mixture of saturated and unsaturated hydrocarbons may be used, e.g. with mothane or ethane alone a temperature of 300—500° C. and a pressure of 600—1000 atmospheres is necessary to produce liquid hydrocarbons, but with a mixture of the unsaturated cracking products of heavy oil and saturated hydrocarbons, the reaction proceeds at 150°—350° C. and 200—400

atmospheres pressure with a commercial useful yield. The actual working conditions, such as

 β -ray velocity, amount and intensity of X-rays, magnitude of the applied electric field, electrical conductivity of the reacting gases, pressure and temperature, must be adjusted according to the end product required. The time of exposure to the action of the rays, the duration of the after effect, and a series of conditions which can be fixed when the end product and initial mixtures are known must also

be taken into account. In most cases, a

mixture of hydrocarbons of various compositions and boiling points is formed, which contains ketones, alcohols etc. These last are formed in quantity if the irradiated gases contain water vapour. The different fractions can be separated by known means such as fractional dis-

tiliation etc.

The vapours of hydrocarbons of low boiling point may be exposed to the action of the rays to form higher or more 60 saturated compounds than the original ones.

The following example illustrates the

process...

Water gas is prepared from coke and purified by known methods. The gas so

obtained is led slowly, continuously and at normal temperature (preferably howtemperature of a 200°—450° C.) through a β -ray apparatus, under the influence of β -rays with a velocity of 200—250.000 kilometres per second. After leaving the apparatus the activated gas is compressed to 800-1000 atmospheres. The length of time for which this pressure is maintained depends 75

a. on the strength of the rays and the time of exposure.

b. on the temperature of the gas during irradiation.

c. on the type of hydrocarbon to be

produced.

hydrocarbons obtained The -(after removal of the CO₂ formed in the synthesis) can be separated by fractional distillation. It is possible, using pure water gas, to produce hydrocarbons of which 60—80% boil below 150° C. With longer irradiation and circulation under increased pressure, the yield of oily hydrocarbons can be raised to 40% or more.

The benzine obtained is mostly of a saturated nature. If small amounts of unsaturated hydrocarbons occur, they can be removed by known methods, such as

sulphuric acid etc.

The process may be shortened by the addition of 10—50% ethylene, acetylene, or other unsaturated hydrocarbon to the water gas. Known catalysts can also be used, and the time of circulation and mag- 100 nitude of the pressure shortened accordingly. Special catalysts may be used, such as radium salts or fluorescent or phosphorescent substances.

It is advisable to cool the reaction 105 chamber when using catalysts, or too great a degree of polymerisation may take place, while too high a temperature may give

rise to cracking products.

It is preferable to use intense radiation 110 and high pressures, dispensing with catalysts and with the necessity of freeing the water gas from catalyst poisons. If catalysts are not used the absence of poison is not so necessary, nor is the presence of 115 nitrogen or water vapour so dangerous, though preferably the water-gas does not contain too much of either. The after effect of the irradiation may be utilised to full effect by circulating the reacting 120 gases under pressure through the apparatus in a cyclical process.

If the oils obtained have too low a viscosity it can be increased if the oil, heated to between 30-150° C. is subjected to 123

further treatment with the rays.

Dated this 29th day of December, 1927.
W. P. THOMPSON & Co.,
12, Church Street, Liverpool, Chartered & Registered Patent Agents.

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COMPLETE SPECIFICATION.

Process for the Synthesis of Liquid Hydrocarbons.

M.Sc. I, HAROLD Edwin Potts, Chartered Patent Agent, of 12, Church Street, Liverpool, in the County of Lancaster, Subject of the King of Great Britain, do hereby declare the nature of this invention which has been communicated to me by Hermann Plauson, a Citizen of Esthonia, of Hagedornstrasse 51, Hamburg 37, Germany, and in what 10 manner the same is to be performed, to be particularly described and ascertained in and by the following statement :-

This invention relates to a process for the synthesis of liquid hydrocarbons of various boiling points from gases such as water gas, and/or gaseous hydrocarbons, such as coke oven gas, natural gas or gases

produced by cracking heavy oils.

According to the present invention liquid hydrocarbons obtained by subjecting mixtures containing gaseous carboncompounds to the action of an intense beam of β rays of velocity exceeding one half the velocity of light or to that of such β rays and the secondary X rays produced therefrom.

An important feature of the invention consists in subjecting the β rays in presence of the reacting gases or not to the action of intense electric and/or rotat-

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ing electromagnetic fields.

Further features of the invention consist in subjecting the activated gases during or especially after irradiation to the action of high pressures and/or temperatures; in the latter case a "ripening" or after reaction occurs. The invention further includes performing the reaction in the presence of catalysts.

Another important feature of the invention is the use of mixtures of saturated and unsaturated hydrocarbons as the initial substances in the synthesis of

liquid hydrocarbons.

The β -rays may be produced by apparatus such as a Coolidge electron tube or that described in my Patent Specification 299,735, or by apparatus constructed on similar principles. In this apparatus the 50 electrons are emitted in large quantities from a surface coated with the clements or oxides of the elements of the rare earth series, heated electrically or otherwise, and enclosed in a highly evacuated chamber. A window of thin metal foil through which the rays can pass freely is provided in the chamber opposite the emitting surface and a reaction chamber is arranged externally to the window.

The β -rays must have sufficiently high

velocities and concentrations. of between 200,000 and 260,000 kilometres per second is desirable and in all cases the velocity preferably exceeds one half the velocity of light. The voltage applied to the β -ray tube to accelerate the rays to this velocity varies between 50,000 and 500,000 volts. For most practical purposes a potential of 250-350,000 volts is sufficient.

The admixture of X-rays may be produced from the action of the β -rays on such substances as glass, quartz, felspar, arranged above the metal window. The action of the β -rays on such substances produces X-radiation, the ionisation caused by the X-rays assisting the action of the \$\beta\-rays on the gases or vapours.

A main anode is arranged opposite to the metal window, and may form one wall of the reaction chamber. This anode is preferably maintained at a high positive potential and the reacting substances pass between the anode and the metal window, so that they are simultaneously subjected to the action of Brays, X-rays, and an intense electric field. An alternating voltage may however be applied to the anode.

A plurality of β-ray tubes may act in 90 parallel on a single reaction chamber, and the gases may be introduced in the direction of, transversely, or in counter

flow to the β -rays.

The power of β -rays to promote chemical 95 reactions may be enhanced by causing the Brays to rotate so that they traverse a spiral or helical path. Such a motion may be caused by allowing the rays to traverse a rotating magnetic field. A 100 rotating magnetic field may be produced by passing three phase alternating current through a system of three coils such as are used to produce a rotating magnetic field in alternating current motors. The I05 combined effect of the rotating magnetic field and the velocity of the electrons forming the rays causes the rays to tra-verse a spiral or helical path.

Catalysts may be used to accelerate the 110 reactions. Suitable catalysts are those known in the heat-pressure synthesis of liquid hydrocarbons from water gas. In the present invention it is possible to increase the yield by circulating the react- 115 ing substances under increased pressures and/or temperatures e.g. to synthesise liquid hydrocarbons from water gas alone. a pressure of 200-1000 atmospheres is desirable; with a mixture of water gas and 120

ethylene or acetylene, the reaction proceeds at ordinary pressures with a 20-30% yield and with increased pressures of 50-200 atmospheres approaches the theoretical yield. If desired, a mixture of saturated and unsaturated hydrocarbons may be used, e.g. with methane or ethane alone a temperature of 300—500° C. and a pressure of 600—1000 atmospheres is necessary to produce liquid hydrocarbons, but with a mixture of the unsaturated cracking products of heavy oil and saturated hydrocarbons, the reaction proceeds at 150°-350° C. and 200-400 atmospheres pressure with a commercially useful yield.

The actual working conditions, such as β -ray velocity, amount and intensity of X-rays, magnitude of the applied electric 20 field, electrical conductivity of the reacting gases, pressure and temperature, must be adjusted according to the end product The time of exposure to the required. action of the rays, the duration of the 25 after effect, and a series of conditions which can be fixed when the end product and initial mixture are known must also be taken into account. In most cases, a mixture of hydrocarbons of various compositions and boiling points is formed, which contains ketones, alcohols etc. These last are formed in quantity if the irradiated gases contain water vapour. The different fractions can be separated by known means such as fractional distillation eto.

The vapours of hydrocarbons of low boiling point may be exposed to the action of the rays to form higher or more saturated compounds than the original ones.

The following example illustrates the

process.

Water gas is prepared from coke and purified by known methods. The gas so obtained is led slowly, continuously and at normal temperature (preferably however at a temperature of from $200^{\circ}-450^{\circ}$ C.) through a β -ray apparatus, 50 under the influence of β -rays with a velocity of 200.000-250.000 kilometres per After leaving the apparatus the activated gas is compressed to 800-1000. atmospheres. The length of time which

this pressure is maintained depends. a. on the strength of the rays and the

time of exposure. b. on the temperature of the gas during

irradiation. c. on the type of hydrocarbon to be

produced.

The hydrocarbons obtained (after removal of the CO₂ formed in the synthusis) can be separated by fractional dis-65 tillation. It is possible, using pure water

gas, to produce hydrocarbons of which 60-80% boil below 150° C. With longer irradiation and circulation under increased pressure, the yield of oily hydrocarbons can be raised to 400% or more.

The benzine obtained is mostly of a If small amounts of saturated nature. unsaturated hydrocarbons occur, they can be removed by known methods, such as sulphurie acid etc.

The process may be shortened by the addition of 10-50% ethylene, acetylene, or other unsaturated hydrocarbon to the water gas.

It is advisable to cool the reaction chamber when using catalysts, or too great a degree of polymerisation may take place. while too high a temperature may give

rise to cracking products.

It is preferable to use intense radiation and high pressures, dispensing with catalysts and with the necessity of freeing the water gas from eatalyst poisons. If catalysts are not used the absence of poison is not so necessary, nor is the presence of nitrogen or water vapour so dangerous. though preferably the water-gas does not contain too much of either. The after effect of the irradiation may be utilised to, full effect by circulating the reacting gases under pressure through the apparatus in a cyclical process.

If the oils obtained have too low a viscosity it can be increased if the oil, heated to between 30-150° C. is subjected to 100 further treatment with the rays.

I am aware of Specification No. 154,213 and claim nothing described or claimed

Having now particularly described and 105 ascertained the nature of the said invention and in what manner the same is to be performed, as communicated to me by my foreign correspondent I declare that .what I claim is :-

1. Process for the production of liquid hydrocarbons, consisting in subjecting mixtures containing gaseous carbon compounds to the action of an intense beam of β rays of velocity exceeding one half the 115 velocity of light, or to that of such β rays and the secondary X-rays derived there-

2. Process as claimed in Claim 1 which consists in subjecting the β rays in 120 presence of the reacting gases or not to the action of intense electric and/or rotating electromagnetic fields.

3. Process as claimed in Claim 1 or 2 in which the activated gases are subjected 125 during or especially after irradiation to the action of high pressure and/or temperature, so that a "ripening" or after reaction occurs.

THE THE

4. Process for the synthesis of liquid 130

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hydrocarbons in which an initial mixture of saturated and unsaturated hydrocarbons is subjected to the polymerising action of β -rays.

5. Process as claimed in any of the preceding claims in which the reaction is performed in the presence of a catalyst or catalysts.

6. Process as claimed in Claim 5 in 10 which the catalytic matter is introduced

in a finely divided form suspended in the reacting gases.

7. Process for the production of liquid or gaseous hydrocarbons, substantially as described.

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Dated this 13th day of February, 1929, W. P. THOMPSON & Co., 12, Church Street, Liverpool, Chartered & Registered Patent Agents.

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