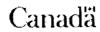
CIPO - Patent - 379224 Page 1 of 2







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(12) Patent:

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(54) ALIPHATIC HYDROCARBON SYNTHESIS

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# SPECIFICATION

TO WHOM IT MAY CONCERN:

Be it known that I, Franz Fischer, of 2, Kaiser Wilhelm Platz, Mülheim-Kuhr, Gormany, Chemist, having invented certain new and useful "Method of increasing yield in the catalytic synthesis of aliphatic hydrocarbons", do hereby declare that the following is a full, clear, and exact description of the same:-

It is known to produce aliphatic hydrocarbons synthetically in the use of catalysts from the oxides of carbon and from hydrogen. It has been found that if a gas mixture of suitable composition - for example a mixture of 1 part carbon monoxide and 2 parts hydrogen - be passed once over a good contact substance at, for example, 190 cand at not too high a velocity, all the carbon monoxide may be converted into hydrocarbons. Thus, as both theory and practice have shown, there are no basic reasons for operating in a number of stages, since the condition of equilibrium at the low temperatures is entirely on the side of the hydrocarbons. Thus the position is not the same as, for example, in the synthetic production of ammonia, where, with the relatively high reaction temperature necessary, for reasons connected with chemical equilibrium, invariably only part of the mixture of nitrogen and hydrogen can be converted into ammonia. In the case referred to it is essential to operate in a number of stages, - during which time the reaction products are removed - the work in practice being effected by circulating over the contact substance.

It has further been found that there is a much greater yield of aliphatic hydrocarbons which contain more than one carbon atom in the molecule (more particularly liquid hydrocarbons which readily volatilise), if the method is carried out in at least two stages and, in between, either the oil or the reaction tater alone, or, in addition, benzine and the hydrocarbons that boil at a still lower temperature, are removed from the gas, care being taken during the early stages, by keeping the temperature low, or by maintaining a high velocity, or by having insufficient hydrogen, or by any combination of these conditions, to ensure that the mixture of carbon monoxide and hydrogen present is not completely consumed but only to the extent of, for example, one-half or two-thirds. By this means the generation of heat within the contact substance is clearly limited, and thus the consumption of the mixture of carcon monoxide and hydrogen is arrested in the direction of the undesirable formation of methane. The process is carried out also in the further stages with the same effect, and only in the last stage is the residue completely converted, where desired, to carbon monoxide and hydrogen.

Thus in the present process, with a view to increasing the yield of aliphatic hydrocarbons which contain more than one carbon atom in the molecule, a step which in itself is unnecessary - namely the division into stages and, in between, the removal of reaction products - is taken with the practical result that there is an increase in yield in the hydrocarbons

required to the extent of 20% and over, up to absost the theoretic limit, such a yield being unattainable in a single stage however carefully the operation oe carried out. Operating in stages, however, without removal of the reaction product has previously been described in connection with the synthetic production of methane from oxides of carbon and hydrogen; there has also been described a method of effecting in the use of catalysts the complete conversion of hydrogen and of oxides of carbon into liquid alighatic hydrocarbons by removing the liquid hydrocarbons produced in one stage but re-converting the undesirable hydrocarbons into carbon monoxide and hydrogen in the use of steam or carbon dioxide or both at a high temperature, and then converting the gas mixture produced into hydrocarbons in the same or in a new contact stage, and so on.

The present invention has nothing in examon with the two methods hereinbefore described. It can be carried out as a circulating process with one or more contact stages, although this is not essential.

### Example I

plus two parts of H<sub>2</sub>) are passed every hour over a contact layer of a length of 90 cms (12 grammes of cobalt) at a temperature of 190°C; the product contains up to 110 grammes of benzine plus oil per cubic metre at normal temperature and pressure.

By using three contact layers of a length of 30 cms (each layer 4 grammes of cobalt) disposed in parallel and passing over each contact layer 4

litres of gas per hour at a temperature of 190° C, the product again contains up to 116 grammes of benzine plus oil per cubic metre of gas at normal temperature and pressure.

over the three contact layers of a length of 30 cms (each layer 4 grammes of cobalt) in succession and, after passing each lager, the reaction products, oil plus benzine plus water, be removed - the temperature of the layers being so selected that in each stage no more than one-third of the original carbon monoxide is converted - the product contains up to 120 grammes of oil plus benzine per cubic metre at normal temperature and pressure.

over two contact layers of a length of 30 cms (each layer 4 grammes of cobalt) disposed parallel to each other and the remainder of the gas is passed over a contact layer of a length of 30 cms disposed to the rear of the other two contact layers, the temperature selected being such that in each layer approximately the same volume of carbon monoxide is converted and then oil, water and benzine are removed from behind each layer, the total product contains up to 130 grammes of benzine plus oil per cubic metre at normal temperature and pressure.

By this method it is possible to work with low temperatures in the early stages, and as a result there is a better yield of liquid hydrocarbons at the expense of the gaseous hydrocarbons which are otherwise produced.

#### Example II

The temperature is not the same in all the

various stages, but is lowest in the first stage and highest in the last. The light benefits and the hydrocarbons which soil at a lower temperature are removed between the several stages, for example by cashing with cold oil, or by the use of active carbon, or by other methods.

## Example III

The same procedure is followed as in Example II except that after each stage all the desired reaction products are removed, the oil being removed by washing with circulating washing oil having a temperature of over 100°C, the water by washing with water, the benzine by washing with cold oil, and the unsaturated gaseous hydrocarbons by washing with suitably concentrated sulphuric acid and the saturated gaseous hydrocarbons by treatment with active carbon.

### Example IV

Complete conversion of the carbon monoxide in the early stages is avoided if an insufficient quantity of hydrogen is present, and after the different stages so much hydrogen, or gas containing a high percentage of hydrogen, be then added to the remaining gas, as will limit the conversion. Only before the last stage is the amount of hydrogen required for complete conversion added. The reaction products are removed between the several stages. The method of working Example IV is applicable when it is desired to secure as large a yield as possible of unsaturated hydrocarbons.

The process hereinbefore inscribed and explained by means of Examples of increasing the yield of aliphatic hydrocarbons which contain more than one carbon atom in the molecule, can be carried out at atmospheric pressure or at higher or lower pressures than atmospheric pressure.

#### I claim:

- 1. A method of increasing the yield in the catalytic synthesis of aliphatic hydrocarbons having more than one carbon atom in the molecule, from carbon monoxide and hydrogen, in which the synthesis is carried out in a number of stages, consisting in effecting partial conversion of the mixture of carbon monoxide and hydrogen in the first stage to the extent of less than 2/3 the available quantity, removing the reaction products and subsequently subjecting the mixture of carbon monoxide and hydrogen to partial conversion in the succeeding stages, each time after separation of the reaction products, and completing the conversion in the final stage.
- 2. A method according to claim 1, wherein the partial conversion is effected by the use of temperatures lying below the optimum conversion temperature.
- 3. A method according to claim 1, wherein the partial conversion is carried out by the use of a content of hydrogen that is deficient with respect to the content of hydrogen with which greater conversion can be secured by carrying out the operation in a single reaction.
- 4. A method according to claim 1, wherein the partial conversion of the mixture or carbon monoxide and hydrogen is effected in the individual stages by the use of high speeds of flow of the gas mixture.
- 5. A method according to claim 1, consisting in partially converting the mixture of carbon monoxide and hydrogen in one of the early stages by the use of a temperature lying below the optimum conversion temperature, removing the condensible reaction products and bringing the conversion of the mixture of carbon monoxide and hydrogen to a conclusion in the further stages by raising



the temperature so that the optimum conversion temperature is attained in the last stage.

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