[Second Edition.]

PATENT SPECIFICATION

Application Date: Aug. 10, 1937. *No. 22010/37.* **500,366**

Complete Specification Left: July 18, 1938.

Complete Specification Accepted: Feb. 8, 1939.

PROVISIONAL SPECIFICATION.

Improvements in Carrying Out Catalytic Reactions in the Gas Phase.

I, GEORGE WILLIAM 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 organised under the Laws of Germany) to be as follows:—

In carrying out catalytic reactions in the gas phase, as for example in hydrogenations, dehydrogenations, oxidations, hydrations and the like, the catalysts used usually subside in their activity after use for more or less long periods so that they must be regenerated or replaced by fresh catalysts. In order to be able to render the regeneration continuous it has already been proposed to lead the catalyst through the reaction chamber, for example on movable substrata or by allowing it to move downwards by reason of its own weight. When working in this way also it is usually unavoidable that the catalyst loses considerably in activity in the reaction chamber and this leads to a diminution in yield.

My foreign correspondents have now found that in catalytic gas reactions the activity of the catalyst can be maintained throughout the whole course of the reaction and an increase in yield be obtained by moving the catalyst through the reaction chamber and allowing the temperature therein to rise in such manner that the output or reaction product remains constant throughout the reaction chamber or does not decrease in an appreciable degree.

In practice, the temperature of the reaction chamber is increased continuously or in stages from the point at which the catalyst enters to the point at which it leaves, care being taken that the movement of the catalyst, which may also be effected continuously or in stages, and the increase in temperature are so selected that the catalyst exerts an action which remains approximately constant. The conditions may readily be determined in a simple manner by preliminary experiment.

The process is suitable for example for the dehydrogenation of hydrocarbons, advantageously those of low molecular weight, as for example for rendering benzines non-knocking under atmospheric or increased pressure; it may also be used in the production of liquid products by the reduction of oxides of carbon; the production of unsaturated hydrocarbons by the incomplete combustion of hydrocarbons with oxygen may also be carried out with advantage by the said process, heat being withdrawn in a suitable manner if necessary.

Other fields of use are for example the hydration of acetylene to form acetaldehyde in the gas phase, and the reaction of acetylene with steam to form acetone.

The process offers special advantages when the catalysts are very short-lived, as for example in the catalytic dehydrogenation of hydrocarbons of low molecular weight at atmospheric pressure. At the high temperatures between 500° and 650° Centigrade to be used, all hitherto known catalysts undergo a comparatively rapid decrease in their activity which renders necessary a frequent regeneration of the catalyst and consequent interruption of the continuous operation. These difficulties are overcome in a simple manner by the present process.

The process will be further described with reference to the accompanying drawing, but the invention is not restricted to the particular arrangement of apparatus shewn.

The catalyst passes from a reservoir V into the vertical reaction chamber in which a definite temperature distribution is maintained by electrical or gas heating H. A transporting device, as for example a conveyor worm F, is provided at the lower end of the reaction chamber and this continuously withdraws a part of the catalyst and moves it to a chamber from which it can be removed through a sluice. The reaction gases enter at the top at *a* into a preheating zone, then meet the fresh, slowly descending

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catalyst and move in the same direction as the latter. The reaction gases leave the reaction chamber somewhat above the conveyor worm F at *b* and are cooled and then further worked up. The reaction gases may also be led in countercurrent. The catalyst need not be withdrawn continuously. An appropriate amount may be withdrawn for example at equal intervals of ½, 1 or 2 hours or more. It is important that the temperature in the reaction chamber should rise proportionately to the decrease in the activity of the catalyst and the increase in temperature and the speed of transportation of the catalyst must therefore be adapted to each other.

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

EXAMPLE 1.

A vertical reaction tube of quartz, constructed similar to that shewn in the drawing, is filled with 120 cubic centimetres of an active carbon which has been prepared by heating lean coal with steam; 2.5 cubic centimetres per hour of the active carbon are withdrawn by a conveyor worm. The same amount of active carbon descends continuously from the reservoir so that the reaction chamber always remains full of the active carbon. Downwards through the reaction chamber technical butane is led at a speed of 24 litres per hour. The reaction chamber is heated externally by three separate electrical heating coils. At the point at which the gas meets the carbon, the latter has a temperature of 530° Centigrade. In the reaction chamber, the temperature of the carbon filling rises and reaches 580° Centigrade at the end thereof. 28 per cent. of the butane passed through are reacted. 80 per cent. of the reacted butane are converted into butylene and propylene. If, however, the whole of the reaction tube be kept at the mean temperature of about 550° Centigrade, only 14 per cent. of the butane are converted of which 70 per cent. consist of butylene and propylene.

EXAMPLE 2.

In the same apparatus as in Example 1, 48 litres per hour of propane are led over an active carbon which has been impregnated with such an amount of aluminium nitrate solution and heated that it contains 3 per cent. of Al_2O_3. The temperature at the inlet is 570° and at the outlet 625° Centigrade. 2.6 cubic centimetres of catalysts are withdrawn per hour; the same amount is added at the top. 33 per cent. of the propane are converted and 60 per cent. of the converted propane are obtained as propylene. This result may be maintained for weeks without trouble.

EXAMPLE 3.

In the apparatus described in Examples 1 and 2 24 litres per hour of isobutane are lead over 120 cubic centimetres of a catalyst consisting of active carbon which has been impregnated with such an amount of iron nitrate and heated that it contains 3.5 per cent. of iron as the oxide. The inlet temperature is 495° and the final temperature 566° Centigrade. 1.7 cubic centimetres of carbon are removed per hour and added again at the top in a fresh form. The final gas contains 14.0 per cent. of isobutylene, 1.6 per cent. of propylene and 0.4 per cent. of ethylene. 22 per cent. of the isobutane are converted by a single passage. The yield of isobutylene amounts to 75 per cent. calculated with reference to the isobutane converted.

Dated this 10th day of August, 1937.

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

COMPLETE SPECIFICATION.

Improvements in Carrying Out Catalytic Reactions in the Gas Phase.

I, GEORGE WILLIAM 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 organised 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:—

In carrying out catalytic reactions in the gas phase, as for example in hydrogenations, dehydrogenations, oxidations, hydrations, and the like, the catalysts used usually subside in their activity after use for more or less long periods so that they must be regenerated or replaced by fresh catalysts. In order to be able to render the regeneration continuous, it has already been proposed to lead the catalyst through the reaction chamber, for example on movable substrata or by allowing it to move downwards by reason of its own weight. When working in this way also it is usually unavoidable that the catalyst loses considerably in activity in the reaction chamber and this leads to a diminution in yield.

My foreign correspondents have now found that in catalytic gas reactions the activity of the catalyst can be maintained throughout the whole course of the reactions and an increase in yield be obtained by moving the catalyst through the reaction chamber and allowing the temperature therein to rise in such manner that the output of reaction product remains constant throughout the reaction chamber or does not decrease in an appreciable degree.

In practice, the temperature of the reaction chamber is increased continuously or in stages from the point at which the catalyst enters to the point at which it leaves, care being taken that the movement of the catalyst. which may also be effected continuously or in stages, and the increase in temperature are so selected that the catalyst exerts an action which remains approximately constant. The conditions may readily be determined in a simple manner by preliminary experiment.

The process is suitable for example for the dehydrogenation of hydrocarbons, advantageously those of low molecular weight, as for example for rendering benzines non-knocking under atmospheric or increased pressure; it may also be used in the production of liquid products by the reduction of oxides of carbon; the production of unsaturated hydrocarbons by the incomplete combustion of hydrocarbons with oxygen may also be carried out with advantage by the said process, heat being withdrawn in a suitable manner if necessary.

Other fields of use are for example the hydration of acetylene to form acetaldehyde in the gas phase, and the reaction of acetylene with steam to form acetone.

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The process will be further described with reference to the drawing accompanying the provisional specification, but the invention is not restricted to the particular arrangement of apparatus shewn.

The catalyst passes from a reservoir V into the vertical reaction chamber in which a definite temperature distribution is maintained by electrical or gas heating H. A transporting device, as for example a conveyor worm F, is provided at the lower end of the reaction chamber and this continuously withdraws a part of the catalyst and moves it to a chamber from which it can be removed through a sluice. The reaction gases enter at the top at *a* into a preheating zone, then meet the fresh, slowly descending catalyst and move in the same direction as the latter. The reaction gases leave the reaction chamber somewhat above the conveyor worm F at *b* and are cooled and then further worked up. The reaction gases may also be led in countercurrent. The catalyst need not be withdrawn continuously. An appropriate amount may be withdrawn for example at equal intervals of ½, 1 or 2 hours or more. It is important that the temperature in the reaction chamber should rise proportionately to the decrease in the activity of the catalyst and the increase in temperature and the speed of transportation of the catalyst must therefore be adapted to each other.

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

EXAMPLE 1.

A vertical reaction tube of quartz, constructed similar to that shewn in the drawing, is filled with 120 cubic centimetres of an active carbon which has been prepared by heating lean coal with steam; 2.5 cubic centimetres per hour of the active carbon are withdrawn by a conveyor worm. The same amount of active carbon descends continuously from the reservoir so that the reaction chamber always remains full of the active carbon. Downwards through the reaction chamber technical butane is led at a speed of 24 litres per hour. The reaction chamber is heated externally by three separate electrical heating coils. At the point at which the gas meets the carbon, the latter has a temperature of 530° Centigrade. In the reaction chamber, the temperature of the carbon filling rises and reaches 580° Centigrade at the end thereof. 28 per cent. of the butane passed through are reacted. 80 per cent. of the reacted butane are converted into butylene and propylene. If, however, the whole of the reaction tube be kept at the mean temperature of about 550° Centigrade, only 14 per cent. of the butane are converted of which 70 per cent. consist of butylene and propylene.

EXAMPLE 2.

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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 carrying out catalytic reactions in the gas phase, such as hydrogenations, dehydrogenations, reduction of oxides of carbon and the like, which comprises moving the catalyst through the reaction zone and raising the temperature therein in the direction of the movement of the catalyst in such a manner that the rate of conversion in the subsequent parts of the reaction zone does not decrease in a substantial degree.

2. The process for carrying out catalytic reactions in the gas phase substantially as described in each of the foregoing Examples.

Dated this 18th day of July, 1938.
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47, Lincoln's Inn Fields, London,
W.C. 2,
Agents.

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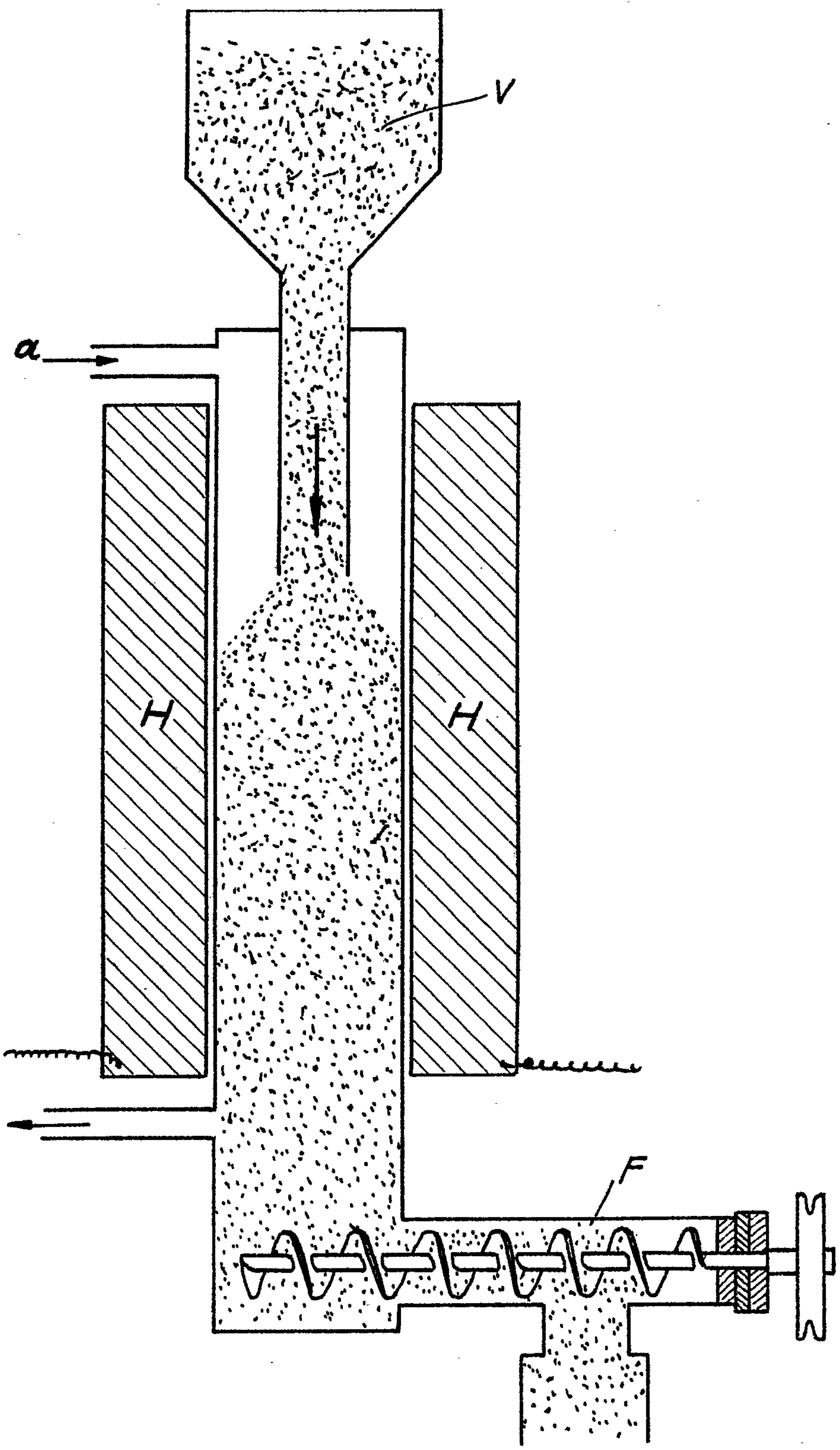
2nd Edition

[This Drawing is a reproduction of the Original on a reduced scale.]



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