#### SPECIFICATION PATENT



Application Date: Aug. 29, 1928. No. 24,839 / 28.

Complete Left: April 15, 1929:

Complete Accepted: Aug. 29, 1929.

PROVISIONAL SPECIFICATION.

# 317,996

## Improvements in the Carrying Out of Gas Reactions.

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)

10 to be as follows: --

When carrying out reactions with gases, or with liquids which are vapourised under conditions at and in the elevated the working temperatures carbon monoxide, 15 of hydrogen, hydrogen sulphide and the like, there is a great strain on the apparatus as regards its capability of resistance especially when working under very great pressures. The 20 material itself too, of which the apparatus is made, may give rise to injurious bye reactions. The attack on the apparatus by the said gases or other gases may be avoided by constructing the apparatus 25 of expensive materials, such as highly alloved special steels and the like, or by coating the apparatus with such materials or other substances which are specially resistant to chemical action. Such 30 materials, however, in addition to being very expensive may in some other way be disadvantageous in use.

My foreign correspondents have now found that the said drawbacks may be 85 avoided, and in particular that less expensive materials may be employed for the purpose required, by adding to the gaseous or under the working conditions, vaporous substances, small quantities of substances 40 which remain liquid under the working conditions, and which are inert towards the materials employed for constructing the apparatus and especially substances which have not too great a solubility as 45 regards the injurious gases, in such a manner that they form a liquid film over the part sof the apparatus with which the reacting substances come into contact at elevated temperatures.

For example in the treatment of middle oil, obtained from corbonaceous materials, tars or heavy mineral oils, in the form of

[Price 3/-]

vapour, with hydrogen under pressure, the hot parts of the apparatus with which the middle oil comes into contact, may be constructed of less expensive materials, as for example of the known and usual slightly alloyed steels, while adding small quantities of oils of high boiling point which remain liquid under the working conditions and which form a thin film on the walls of the apparatus. This takes place for example when the gas and the middle oil to which a non-volatile oil has been added, stream through a coil from the top to the bottom. The middle oil is vapourised whilst the added oil covers the wells of the apparatus with a film of liquid. Conversely the products may be caused to stream through the coil from the bottom to the top with a greater velocity so that the non-volatile oil is carried along with the stream and forms a film of liquid on the walls of the apparatus. In larger vessels, as for example, high pressure reaction vessels, the film may also be produced by allowing oil to trickle round in a thin layer.

The result obtained according to this invention is remarkable, since hydrogen for example, under high pressure, say at 200 atmospheres and above 400° Centigrade produces in so short a time such marked changes in the structure of ordinary steel and the like to which only very small quantities of alloy metals have been added, that its use has hitherto been found to be of no service. Oils of high boiling point such as anthracene oil and the like, or molten substances such as paraffin wax or the like, may be employed according to this invention.

The invention herein described may be successfully applied to other gas reactions such as for example the synthesis of methanol, the synthesis of higher alcohols

from oxides of carbon and reducing gases, and other general reactions in which gases containing carbon monoxide are made use 100

The following Example will further illustrate the nature of the said invention but the invention is not limited thereto. EXAMPLE.

Hydrogen together with 1 kilogram of

middle oil, of a boiling point range of from 200° to 825° Centigrade, per cubic metre and containing 5 per cent. of a parafim of high boiling point are passed through a high pressure coil of Siemens Martin steel at 470° Centigrade and at 200 atmospheses pressure in such a way that the oil flows through the coil from the

top to the bottom. A micro-photograph of the steel shews no change in its structure after use.

Dated this 29th day of August, 1928.

JOHNSONS & WILLCON,

47, Lincoln's Inn Fields, London,

W.C. 2,

Agents.

#### COMPLETE SPECIFICATION:

### Improvements in the Carrying Cut of Gas Reactions.

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

When carrying out reactions in the gaseous phase, namely reactions with gases, or with liquids which are vapourised under the working conditions at elevated temperatures and particularly in the presence of gases which attack the material of the apparatus, such as hydrogen, carbon monoxide, or hydrogen sulphide and the like, there is a great strain on the apparatus as regards its capability of resistance, especially when working under very high pressures. The material itself too, of which the apparatus is made, may give rise to injurious bye reactions. attack on the material of which the sair faces of the apparatus are constructed by the said guses or other gases may be avoided by constructing the apparatus of expensive materials, such as highly alloyed special steels and the like, or by coating the apparatus with such materials or other substances which are specially resistant to chemical action. Such materials, however, in addition to being very expensive may in some other way be disadvantageous in tise;

My foreign correspondents have now found that the said drawbacks can be avoided, and in particular that less expensive materials can be employed for the purpose required by operating in the presence of small quantities of substances which remain liquid under the working condition, and which are inert towards the materials employed for constructing the apparatus, and especially substances

the materials employed for constructing 60 the apparatus, and especially substances which have no great solibility as regards

the injurious gases, in such a manner that they form a liquid film on the parts of the apparatus with which the reacting substances come into contact at elevated temperatures. This may be effected, for example, by adding the said substances remaining liquid to the gaseous or, under the working conditions, vaporous substances under treatment.

Thus in the destructive hydrogenation of carbonaceous materials, in the gaseous phase, for example, in the treatment of middle oil, obtained from curbonaceous materials, tars or heavy mineral oils, in the form of vapour, with hydrogen under pressure, the hot parts of the apparatus with which the middle oil comes into contact, may be constructed of less expensive materials, as, for example, of the known and usual slightly alloyed steels, while adding small quantities of our of high boiling point which remain liquid under the working conditions and which form a thin film on the walls of the apparatus. This takes place, for example, when the gas and the middle oil to which nonvolatile oil has been added, stream through the apparatus, such as a coil from the top to the bottom. The middle oil is vapourised whilst the added oil covers the walls of the apparatus with a film of liquid. Conversely the products may be caused to stream through the coil or the like from the bottom to the top with a greater velocity so that the non-volatile oil is carried along with the stream and forms a film of liquid on the walls of the apparatus. In larger vessels, as, for example, high pressure reaction vessels, 100 the film may also be produced by allowing oil to trickle along the wall in a thin layer. The protective liquid is preferably admitted at the top of the apparatus, and allowed to flow downwards over the 105 surfaces thereof.

The result obtained according to this invention is remarkable, since hydrogen for example, under high pressure, say at 200 atmospheres and above 400° Centi-110 grade produces in so short a time such

75

marked changes in the structure of ordinary steel and the like to which only very small quantities of alloy metals have been added, that the use of these steels bas hitherto been found to be of no service. Oils of high boiling point, such as anthracene oil and the like, or molten substances, such as paraffin wax or the like, may be employed according to this invention. The process according to the present invention may be carried out under any desired pressure, for example, reduced, atmospheric or clevated pressure.

The invention herein described may be successfully applied to other reactions carried out in the gaseous phase, as for example the synthesis of methanol, the synthesis of higher alcohols and other oxygen-containing organic compounds from exides of carbon and reducing gases, and other reactions in which gases containing carbon monoxide are made use of. In these cases oxygen-containing organic compounds of high-molecular weight may, for example, be employed as the protective liquid.

The following Example will further illustrate how the said invention may be carried out in practice, but the invention

30 is not limited thereto.

Hydrogen together with 1 kilogram, per rubic metre, of middle oil, of a boiling point range of from 200° to 325° Centistration of the point range of from 200° to 325° Centistration of the property of the point, is passed through a high pressure coil of Siemens-Martin steel at about 470° Centigrade and at about 200 atmospheres pressure in such 40° a way that the oil flows through the coil

from the top to the bottom. A microphotograph of the steel shews no change in the structure after use.

If the reaction be carried out without the said addition of paraffin wax, a change in the structure of the materials of the walls is observed resulting in a decrease in their solidity.

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. In reactions carried out in the gaseous phase at elevated temperatures and particularly in the presence of gases which attack the material of the apparatus, such as hydrogen, carbon monoxide or hydrogen sulphide and the like, the step of operating in the presence of small quantities of substances which remain liquid under the working conditions, and which are inert towards the materials employed for constructing the apparatus, in such a manner that they form a liquid film on the parts of the apparatus with which the reacting substances come into contact at elevated temperatures.

2. The process for carrying out reactions in the gaseous phase substantially as described in the foregoing Example.

3. The products from reactions carried out in the gaseous phase when obtained in accordance with the preceding claiming clauses.

Dated this 15th day of April, 1939.
J. Y. & G. W. JOHNSON,
47, Lincoln's Inn Fields, London,
W.C. 2,
Agents.

Charles (1966) Table Sylvanian

Ablagdon: Printed for His Majesty's Stationery Office, by Burgess & Son. [Wt. 51A.—125/12/1929.]