PATENT SPECIFICATION

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PROVISIONAL SPECIFICATION

Improvements in and Apparatus for the Removal of Foreign Gases from the Circulating Gases of Catalytic Pressure Reactions

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:—

Gas mixtures which have been subjected to catalytic pressure reactions, as for example the ammonia synthesis, methanol synthesis or destructive hydrogenation, frequently contain, in addition to the reactants, more or less large amounts of other gaseous constituents which do not take part in the reaction, as for example methane, argon, carbon dioxide or the like. In some cases these attendant gases are contained in the initial gases from the start and some of them may first be formed in the reaction by reason of side reactions. These foreign gases become enriched in the reaction gases led in a cycle and must therefore be removed continuously or periodically because otherwise they unfavourably affect the yield by reason of the increasing diminution in the partial pressure of the reactants. If the foreign gases be removed by continuously withdrawing a part of the gas mixture from the cycle, for example to such an extent that the amount of foreign gases contained therein is the same as the amount thereof introduced during the same time with the fresh gas or by conversion, there is also naturally a waste of the valuable gas constituents at the same time which may be quite considerable. In order to reduce this waste, the procedure may also be that the circulating gases are treated with liquids which have a good solvent power for the foreign gases but which do not dissolve the reactants or only dissolve them to a slight extent. High-boiling oil fractions have been proposed as such solvents for the removal of foreign gases from the circulating gases in the ammonia synthesis. When using such a substance foreign to the operation, the danger of the introduction of catalyst poisons into the gas cycle cannot always be avoided with certainty. The reaction product which is liquid or which has been liquefied by cooling and which has been formed in the reaction may itself be used as solvent for the foreign gases, preferably in a cycle by freeing the liquid reaction product from the absorbed foreign gases by releasing the pressure and then using it again. In this case, however, considerable amounts of the reactants are also removed from the cycle together with the foreign gases and are therefore withdrawn from their proper purpose.

My foreign correspondents have now found that the removal of foreign gases from the circulating gases of catalytic pressure reactions by means of the liquid reaction product can be considerably improved by introducing the fresh gas serving to replenish the circulating gas only into a part of the circulating system advantageously into the larger part of a circulating system consisting in known manner of single cycles arranged in parallel, and only bringing the gas mixture circulating at the end of the other part of the circulating system into contact with the separated liquid reaction product for the purpose of washing out the foreign gases. By working in this way, the foreign gases become enriched in the part of the circulating system not charged with fresh gas so that the highest content of foreign gases occurs in the end cycle. Since in this way the partial pressure of the foreign gases is greatly increased, correspondingly larger amounts thereof dissolve in the treatment with the liquid reaction product. Furthermore those amounts of the reactants which are absorbed by the liquid reaction product during its separation in each single cycle, are for the most part given up again in exchange for the foreign gases during its treatment with the gas mixture in the end cycle. With a given content of fresh gas to foreign gas, the content of foreign gas in the end cycle is continuously regulated so that equally large amounts of foreign gas are

[*Price* 1/-]

Price 4s 6d

removed as enter with the fresh gas during the same time.

It is not necessary to use the liquid reaction product in circulation and to release it from pressure before each re-employment, but it is only necessary to use it and to release it from pressure once after it has been laden strongly with foreign gases and consequently to a correspondingly lesser extent with the valuable gases. There are thus obtained very high yields with great average efficiency of the catalyst chamber and good utilisation of the whole plant.

The nature of the invention will be further described with reference to the accompanying drawing in which is shown by way of example an arrangement of apparatus according to this invention but the invention is not restricted to the particular arrangement shewn. *a* are circulating pumps, *b* are catalyst chambers, *c* are the coolers and *d* are the separators of a series of cycle devices arranged in parallel, of which any number may be used. The fresh gas is not introduced into all the single cycles, but at *e* into only two of three, or into five of seven of the single cycles present. In parallel to the last or end cycle in the other part of the whole system there is arranged by means of gas pipes *f* a washing tower *g* which is preferably provided with filler bodies. The gas mixture of the final cycle is led through this washing tower *g* upwards. In counter-current thereto there flows over the filler bodies the liquid reaction product collected from the separators *d* and led to the upper end of the washing tower *g* through a pipe *h*. It absorbs the single components of the gas mixture in an amount depending on their partial pressures. At the lower end of the washing tower *g* it is released from pressure through a pipe *i* into a reservoir or intermediate container *k*. In the latter a pressure is maintained which corresponds to the pressure of the liquid product at the prevailing external temperature or which is only slightly above the said pressure. The gas mixture escaping from the liquid and which consists mainly of the foreign gases is led away through a pipe *l*. The washing tower *g* may also be interposed directly in the last cycle.

When necessary or desirable, additional amounts of the end cycle gas mixture may be removed through a valve *p* whereby the content of foreign gases can be still further reduced or kept at any desired value.

When using the process for the ammonia synthesis, the temperature of the liquid ammonia in the washing tower preferably amounts to from about 20° to 30° Centigrade, to which it must be heated in some cases. The liquid ammonia has the property of having a solvent power for gas which increases with increasing temperature. The heating up of the liquid ammonia obtained in the separators at a low temperature can be effected in the washing tower itself directly by the circulating gas. In this case the end cycle gas to be washed is preferably branched off before or behind the cooler *c*. The gas leaving the washing tower may be returned to the cycle at any other suitable place instead of directly before the suction side of the circulating pumps.

The apparatus required for the process are very simple. The process may be carried out without the additional use of special machine parts, such as gas compressors, pressure pumps and the like and with merely the utilisation of pressure differences present in any cycle plant. If necessary the said pressure difference may be increased by inserting dam discs or regulating valves. According to this invention there may also be worked up with advantage synthesis gases which have a comparatively high content of methane and other foreign gases by reason of their preparation from bituminous fuels, such as brown coals or lignite small coke, or of their recovery from coke-oven gases.

Dated this 16th day of July, 1937.
J. Y. & G. W. JOHNSON,
47, Lincoln's Inn Fields, London, W.C.2,
Agents.

COMPLETE SPECIFICATION

Improvements in and Apparatus for the Removal of Foreign Gases from the Circulating Gases of Catalytic Pressure Reactions

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:—

Gas mixtures which have been subjected to catalytic pressure reactions, as for example the ammonia synthesis, methanol synthesis or destructive hydrogenation, frequently contain, in addition to the reactants, more or less large amounts of other gaseous constituents, which do not take part in the reaction, as for example methane, argon, carbon dioxide or the like. In some cases these attendant gases are contained in the initial gases from the start and some of them may first be formed in the reaction by reason of side reactions. These foreign gases become enriched in the reaction gases led in a cycle and must therefore be removed continuously or periodically because otherwise they unfavourably affect the yield by reason of the increasing diminution in the partial pressure of the reactants. If the foreign gases be removed by continuously withdrawing a part of the gas mixture from the cycle, for example to such an extent that the amount of foreign gases contained therein is the same as the amount thereof introduced during the same time with the fresh gas or by conversion, there is also naturally a waste of the valuable gas constituents at the same time which may be quite considerable. In order to reduce this waste, the procedure may also be that the circulating gases are treated with liquids which have a good solvent power for the foreign gases but which do not dissolve the reactants or only dissolve them to a slight extent. High-boiling oil fractions have been proposed as such solvents for the removal of foreign gases from the circulating gases in the ammonia synthesis. When using such a substance foreign to the operation, the danger of the introduction of catalyst poisons into the gas cycle cannot always be avoided with certainty. The reaction product which is liquid or which has been liquefied by cooling and which has been formed in the reaction may itself be used as solvent for the foreign gases, preferably in a cycle by freeing the liquid reaction product from the absorbed foreign gases by releasing the pressure and then using it again. In this case, however, considerable amounts of the reactants are also removed from the cycle together with the foreign gases and are therefore withdrawn from their proper purpose.

My foreign correspondents have now found that the removal of foreign gases from the circulating gases of catalytic pressure reactions by means of the liquid reaction product can be considerably improved in such circulating systems which consist of single cycles arranged in parallel, each single cycle comprising the essential devices such as catalyst chamber, circulating pump, cooler and separator. According to this invention the fresh gas serving to replenish the circulating gas is introduced only into a part, advantageously into the larger part of the single cycles arranged in parallel, and the gas mixture circulating at the end of the other part of the circulating system is brought into contact with the separated liquid reaction product for the purpose of washing out the foreign gases. When working in this way, the foreign gases continuously wander from the cycles charged with fresh gas into the other part of the system which is not charged with fresh gas. In the latter part the foreign gases become enriched, and the highest content of foreign gases occurs in the end cycle. Since in this way the partial pressure of the foreign gases is greatly increased, correspondingly larger amounts thereof are dissolved when being treated with the liquid reaction product. Furthermore those amounts of the reactants which are absorbed by the liquid reaction product during its separation in each single cycle, are for the most part given up again in exchange for the foreign gases during the treatment of the gas mixture of the end cycle. With a given foreign gas content of the fresh gas, the content of foreign gas in the end cycle is continuously regulated so that equally large amounts of foreign gas are removed as enter with the fresh gas during the same time.

It is not necessary to use the liquid reaction product in circulation and to release it from pressure before each re-employment, but it is only necessary to use it and to release it from pressure once after it has been laden strongly with foreign gases and consequently to a correspondingly lesser extent with the valuable gases. There are thus obtained very high yields with great average efficiency of the catalyst chamber and a good utilisation of the whole plant.

The nature of the invention will be further described with reference to the drawing accompanying the provisional specification in which is shewn by way of example an arrangement of apparatus according to this invention but the invention is not restricted to the particular arrangement shewn.

a are circulating pumps, *b* are catalyst chambers, *c* are the coolers and *d* are the separators of a series of cycle devices

arranged in parallel, of which any number may be used. The fresh gas is not introduced into all the single cycles, but at *e* into only two of three, or into five of seven of the single cycles present. In parallel to the last or end cycle in the other part of the whole system there is arranged by means of gas pipes *f* a washing tower *g* which is preferably provided with filler bodies. The gas mixture of the end cycle is led through this washing tower *g* upwards. In counter-current thereto there flows over the filler bodies the liquid reaction product collected from the separators *d* and led to the upper end of the washing tower *g* through a pipe *h*. It absorbs the single components of the gas mixture in an amount depending on their partial pressures. At the lower end of the washing tower *g* it is released from pressure through a pipe *i* into a reservoir or intermediate container *k*. In the latter a pressure is maintained which corresponds to the pressure of the liquid product at the prevailing external temperature or which is only slightly above the said pressure. The gas mixture escaping from the liquid and which consists mainly of the foreign gases is led away through a pipe *l*. The washing tower *g* may also be interposed directly in the end cycle.

When necessary or desirable, additional amounts of the end cycle gas mixture may be removed through a valve *p* whereby the content of foreign gases can be still further reduced or kept at any desired value.

When using the process for the ammonia synthesis, the temperature of the liquid ammonia in the washing tower preferably amounts to from about 20° to 30° Centigrade, to which it must be heated in some cases. The liquid ammonia has the property of having a solvent power for gas which increases with increasing temperature. The heating up of the liquid ammonia obtained in the separators at a low temperature can be effected in the washing tower itself directly by the circulating gas. In this case the end cycle gas to be washed is preferably branched off before or behind the cooler *c*. The gas leaving the washing tower may be returned to the cycle at any other suitable place instead of directly before the suction side of the circulating pumps.

The apparatus required for the process are very simple. The process may be carried out without the additional use of special machine parts, such as gas compressors, pressure pumps and the like and with merely the utilisation of pressure differences present in any cycle plant. If necessary the said pressure difference may be increased by inserting dam discs or regulating valves. According to this invention there may also be worked up with advantage synthesis gases which have a comparatively high content of methane and other foreign gases by reason of their preparation from bituminous fuels, such as brown coals or lignite small coke, or of their recovery from coke-oven gases.

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. Improvements in the removal of foreign gases from the circulating gases of catalytic pressure reactions performed in circulating systems consisting of single cycles arranged in parallel, by treating the gases with liquid reaction product and replenishing the circulating gas by fresh gas, which improvements consist in introducing the fresh gas serving to replenish the circulating gas, only into a part of the single cycles, but not into the end cycle, and bringing the gas mixture circulating at the end of the system into contact with the separated liquid reaction product.

2. In the process as claimed in claim 1, employing liquid ammonia of a temperature of about 20° to 30° Centigrade for treating the circulating gases of the ammonia synthesis in the said manner.

3. A process and an apparatus for the removal of foreign gases from the circulating gases of catalytic pressure reactions substantially as described with reference to the drawing accompanying the Provisional specification.

Dated this 18th day of July, 1938.

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

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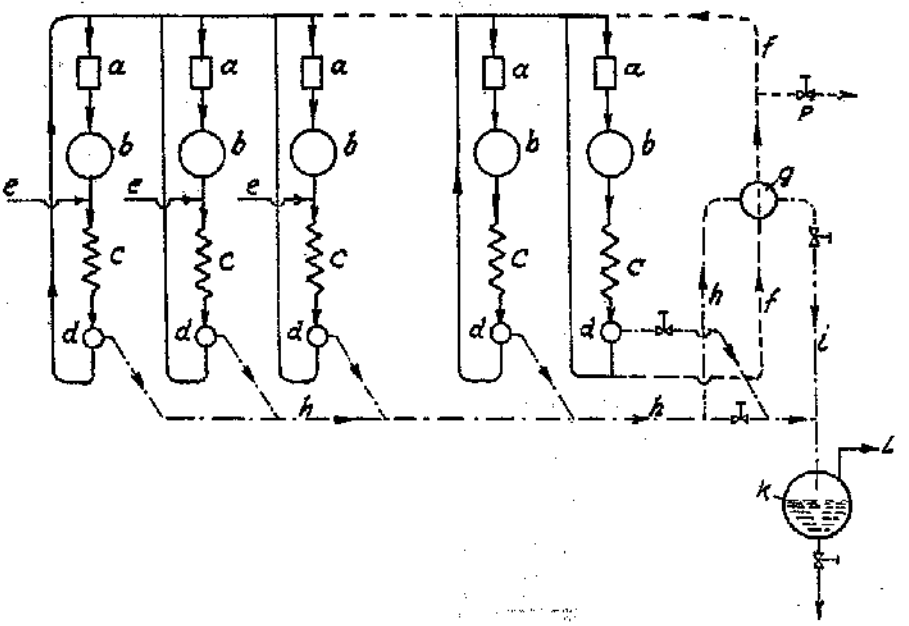

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