[Second Edition.]

PATENT SPECIFICATION

Application Date: Jan. 30, 1934. No. 3170/34.

Complete Specification Left: Jan. 30, 1935.

Complete Specification Accepted: July 30, 1935.

432,574

8 23

PROVISIONAL SPECIFICATION.

Improvements in and relating to the Carrying Out of Chemical Reactions with Circulating Gases.

We, KENNETH GORDON, a British Subject, of Norton Hall, The Green, Norton-on-Tees, County Durham, and IMPERIAL CHEMICAL INDUSTRIES LIMITED, a British Company, of Imperial Chemical House, Millbank, London, S.W.1, do hereby declare the nature of this invention to be as follows:—

This invention relates to the carrying out of chemical reactions with circulating gases, especially under high pressure of the order generally used in the synthetic ammonia industry.

In such reactions it is sometimes necessary to remove a proportion of certain of the gases in order to maintain steady operating conditions, and this can be done by purging a fraction of the circulating gases, by treating the circulating gases with a suitable liquid or solid absorbent, or by condensing out the gases it is desired to remove. Naturally any such step must be taken at a suitable point, having regard to the arrangements made for separating the reaction products. As a rule, it is convenient to treat the circulating gases for the removal of undesired constituents at a point subsequent to the removal of the reaction products.

We have found that a convenient and economic method of removing a proportion of one or more of the constituents of the gases, is to pass the circulating gases in rapid co-current flow with a suitable liquid absorbent, for example by passing the gases and the liquid through a coil of pipe of sufficient length to ensure the desired degree of absorption. The exit liquid may be regenerated by driving off the absorbed gas and used again in the process, or it may be recirculated continuously and a certain fraction of it withdrawn for regeneration, the regenerated liquid being returned to the system. Fresh liquid is added as required to make up for any losses.

Our improved method of removing a proportion of the gases leads to an increased absorption per unit volume of liquid compared with that obtainable in a counter-current washing process, provided that the proportion of gas removed is relatively small so that the partial pressures of the constituent gases are not materially changed. We can also use higher gas velocities than in the counter-current process, since in the latter case the gas velocity must be kept low in order to permit of the proper distribution of liquid and to prevent liquid being entrained by the gas and carried out of the top of the washer.

As a result of the increased absorption per unit volume of absorbent liquid our method requires a smaller volume of circulating liquid, and since we can use high rates of gas flow the size of plant required is materially reduced compared with that required to operate a counter-current absorption process. Also, owing to the smaller volume of absorbent liquid, the losses of valuable gases, e.g. hydrogen, which are unavoidably dissolved by the liquid but which it is not desired to remove from the system, are correspondingly reduced.

Our invention may be applied to the treatment of the circulating gases, after removal of the chief reaction products, in processes for the synthesis of ammonia from nitrogen and hydrogen, or in processes for the synthesis of methanol from carbon monoxide and hydrogen, or in processes for the destructive hydrogenation of carbonaceous materials in the liquid or vapour phase. In the synthesis of ammonia the gases to be removed are methane and inert gases such as argon, which are introduced with the make-up gases and which tend to accumulate in the system. In the synthesis of methanol the gases to be removed are mainly nitrogen introduced with the make-up gases and methane, which is partly introduced with the make-up gases and partly produced by a side reaction. In the destructive hydrogenation of carbonaceous materials the gases to be removed are mainly methane and ethane, which are produced during the course of the reaction, and nitrogen introduced with the make-up hydrogen. Instead of treating the whole of the circulating gases, we may by-pass a portion of the gases and treat the by-passed gases as

[*Price* 1s.]

Price 4s 6d.

described before returning them to the circulating system. In this way the system is made more flexible as the gas and liquid rates are independently controllable.

Our invention may also be applied to the removal of gaseous or vaporous reaction products in cases where it is desired to effect the reaction with the initial presence of a substantial proportion of the reaction products and only to remove from the system the amounts of reaction products corresponding to the reactants used up.

By way of example we will describe the application of the invention to the removal of methane, ethane and nitrogen from the circulating gases of a destructive hydrogenation plant operating under a pressure of 200 atmospheres. The gases circulated through a converter, where they were brought into contact with the carbonaceous material at a raised temperature, then through a cooled catch-pot, where the condensible products were separated from the gases, and then through a gas washer supplied with kerosene as the absorbent liquid for removing methane, ethane and nitrogen. The rate of gas circulation was 5000 cubic metres per hour, referring to the total gas measured at normal temperature and pressure. The gas entering the converter contained 10 per cent. by volume of nitrogen and 5 per cent. of methane and ethane taken together, and in order to keep the composition of the circulating gas constant at this point it was necessary to remove nitrogen at the rate of 12 cubic metres per hour and methane and ethane at the rate of 20 cubic metres per hour. The gases leaving the catch-pot were passed upwardly through a washer consisting of a pipe 17 cm. in diameter and 4 metres in length, kerosene being injected at the base at the rate of 5.3 cubic metres per hour, and the required amounts of nitrogen, methane and ethane were thus removed from the gases.

By way of comparison it may be mentioned that in a counter-current washing process, using the maximum gas velocity consistent with proper distribution of the kerosene, four lengths of pipe each 17 cm. in diameter and 12 metres in length were necessary to fulfil the same duty as the single pipe 4 metres in length mentioned above. The gas was passed in parallel streams through the four pipes and kerosene was injected at the total rate of 6.3 cubic metres per hour. Thus in the co-current process a much smaller apparatus was required and one cubic metre per hour less of kerosene than in the counter-current process, corresponding to a decreased loss of 12 cubic metres of dissolved hydrogen.

In the co-current process the absorption efficiency was 95 per cent. compared with 80 per cent. in the case of the counter-current process.

Dated the 29th day of January, 1934.
E. A. BINGEN,
Imperial Chemical House,
Millbank, London, S.W.1.
Solicitor for the Applicants.

COMPLETE SPECIFICATION.

Improvements in and relating to the Carrying Out of Chemical Reactions with Circulating Gases.

We, KENNETH GORDON, a British Subject, of Norton Hall, The Green, Norton-on-Tees, County Durham, and IMPERIAL CHEMICAL INDUSTRIES LIMITED, a British Company, of Imperial Chemical House, Millbank, London, S.W.1, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

This invention relates to the carrying out of chemical reactions with circulating gases, especially under high pressure of the order generally used in the synthetic ammonia industry.

In such reactions it is sometimes necessary to remove a proportion of certain of the gases in order to maintain steady operating conditions, and this can be done by purging a fraction of the circulating gases, by treating the circulating gases with a suitable liquid or solid absorbent, or by condensing out the gases it is desired to remove. Naturally any such step must be taken at a suitable point, having regard to the arrangements made for separating the reaction products. As a rule, it is convenient to treat the circulating gases for the removal of undesired constituents at a point subsequent to the removal of the reaction products.

We have found that a convenient and economic method of removing a proportion of one or more of the constituents of the gases, is to pass the circulating gases in rapid co-current flow with a suitable liquid absorbent, for example by passing the gases and the liquid, preferably under pressure, through a pipe or a coil of pipe of sufficient length to ensure the desired degree of absorption. The exit liquid may be regenerated by driving off the absorbed gas (for example by releasing the pressure) and used again in the process, or it may be recirculated continuously and a certain fraction of it withdrawn for regeneration, the regenerated liquid being returned to the system. Fresh liquid is added as required to make up for any losses.

Our improved method of removing a proportion of the gases leads to an increased absorption per unit volume of liquid compared with that obtainable in a counter-current washing process, provided that the proportion of gas removed is relatively small so that the partial pressures of the constituent gases are not materially changed. We can also use higher gas velocities than in the counter-current process, since in the latter case the gas velocity must be kept low in order to permit of the proper flow of liquid down the tower and to prevent liquid being entrained by the gas and carried out of the top of the washer.

As result of the increased absorption per unit volume of absorbent liquid our method requires a smaller volume of circulating liquid, and since we can use high rates of gas flow the size of plant required is materially reduced compared with that required to operate a counter-current absorption process. Also, owing to the smaller volume of absorbent liquid, the losses of valuable gases, e.g. hydrogen, which are unavoidably dissolved by the liquid but which it is not desired to remove from the system, are correspondingly reduced.

Our invention may be applied to the treatment of the circulating gases, after removal of the chief reaction products, in processes for the synthesis of ammonia from nitrogen and hydrogen, or in processes for the synthesis of methanol from carbon monoxide and hydrogen, or in processes for the destructive hydrogenation of carbonaceous materials in the liquid or vapour phase. In the synthesis of ammonia the gases to be removed are methane and inert gases such as argon, which are introduced with the make-up gases and which tend to accumulate in the system. In the synthesis of methanol the gases to be removed are mainly nitrogen introduced with the make-up gases and methane, which is partly introduced with the make-up gases and partly produced by a side reaction. In the destructive hydrogenation of carbonaceous materials the gases to be removed are mainly methane and ethane, which are produced during the course of the reaction, and nitrogen introduced with the make-up hydrogen. Instead of treating the whole of the circulating gases, we may by-pass a portion of the gases and treat the by-passed gases as described before returning them to the circulating system. In this way the system is made more flexible as the gas and liquid rates are independently controllable.

Our invention may also be applied to the removal of gaseous or vaporous reaction products in cases where it is desired to effect the reaction with the initial presence of a substantial proportion of the reaction products and only to remove from the system the amounts of reaction products corresponding to the reactants used up.

By way of example we will describe the application of the invention to the removal of methane, ethane and nitrogen from the circulating gases of a destructive hydrogenation plant operating under a pressure of 200 atmospheres. The gases circulated through a converter, where they were brought into contact with the carbonaceous material at a raised temperature, then through a cooled catch-pot, where the condensible products were separated from the gases, and then through a gas washer supplied with kerosene or the like as the absorbent liquid for removing methane, ethane and nitrogen. The rate of gas circulation was 500 cubic metres per hour, referring to the total gas measured at normal temperature and pressure. The gas entering the converter contained 10 per cent. by volume of nitrogen and 5 per cent. of methane and ethane taken together, and in order to keep the composition of the circulating gas constant at this point it was necessary to remove nitrogen at the rate of 12 cubic metres per hour and methane and ethane at the rate of 20 cubic metres per hour. The gases leaving the catch-pot were passed upwardly through a washer, consisting of a pipe 17 cm. in diameter and 4 metres in length, kerosene being injected at the base at the rate of 5.3 cubic metres per hour, and the required amounts of nitrogen, methane and ethane were thus removed from the gases.

By way of comparison it may be mentioned that in a counter-current washing process, using the maximum gas velocity which permits the flow of the kerosene down the tower without kerosene being entrained by the gas and being carried out of the top of the tower, four lengths of pipe each 17 cm. in diameter and 12 metres in length were necessary to fulfil the same duty as the single pipe 4 metres in length mentioned above. The gas was passed in parallel streams upwardly through the four pipes and kerosene was injected downwardly at the total rate of 6.9 cubic metres per hour. Thus in the co-current process a much smaller apparatus was required and one cubic metre per hour less of kerosene than in the counter-current process, corresponding to a decreased loss of 12 cubic metres of dissolved hydrogen.

In the co-current process the absorption efficiency was 95 per cent. compared with 80 per cent. in the case of the counter-current process.

In British Specification 331,916 it has been proposed to recover the reaction products from the gases and vapours obtained in the destructive hydrogenation of distillable carbonaceous materials by condensing the vapours and simultaneously bringing the gases and vapours into contact with a washing oil which may be introduced into the condenser in co-current with the said gases and vapours. In the said specification it is stated that the uncondensed gases are freed to a great extent or practically completely from the hydrocarbons, whereas according to the present invention only a small proportion of the gases is removed in the course of a single passage through the scrubbing zone.

Having now particularly described and ascertained the nature of our said invention and in what manner the same is to be performed, we declare that what we claim is:—

1) A process for carrying out chemical reactions with circulating gases, in which a proportion of one or more of the constituents of the gases is removed by passing the circulating gases, or a portion of them, in rapid co-current flow with a suitable liquid absorbent under such conditions that the proportion of gas removed is relatively small, and that the partial pressures of the constituent gases are not thereby materially changed, the liquid absorbent being subsequently regenerated and returned to the system.

2) A process as claimed in Claim 1, in which the gases and the liquid are passed through a pipe or a coil of pipe of sufficient length to ensure the desired degree of absorption.

3) A process as claimed in Claim 1 or 2, in which the gases and the liquid are passed upwardly.

4) A process as claimed in Claim 1, 2 or 3, in which the gases are treated under high pressure.

5) A process as claimed in Claim 1, 2, 3 or 4, in which the liquid absorbent is recirculated continuously and a certain fraction of it is withdrawn for regeneration, the regenerated liquid being returned to the system.

6) A process as claimed in any of Claims 1—5, in which the circulating gases are those of a destructive hydrogenation plant operated under high pressure and the liquid absorbent is kerosene or the like.

Dated the 30th day of January, 1935.

E. A. BINGEN,
Imperial Chemical House,
Millbank, London, S.W. 1,
Solicitor for the Applicants.

Abingdon: Printed for His Majesty's Stationery Office, by Burgess & Son.
[Wt. 8023.—50/11/1935.