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METHOD OF SEPARATING THE CONSTITUENTS OF GASEOUS MIXTURES

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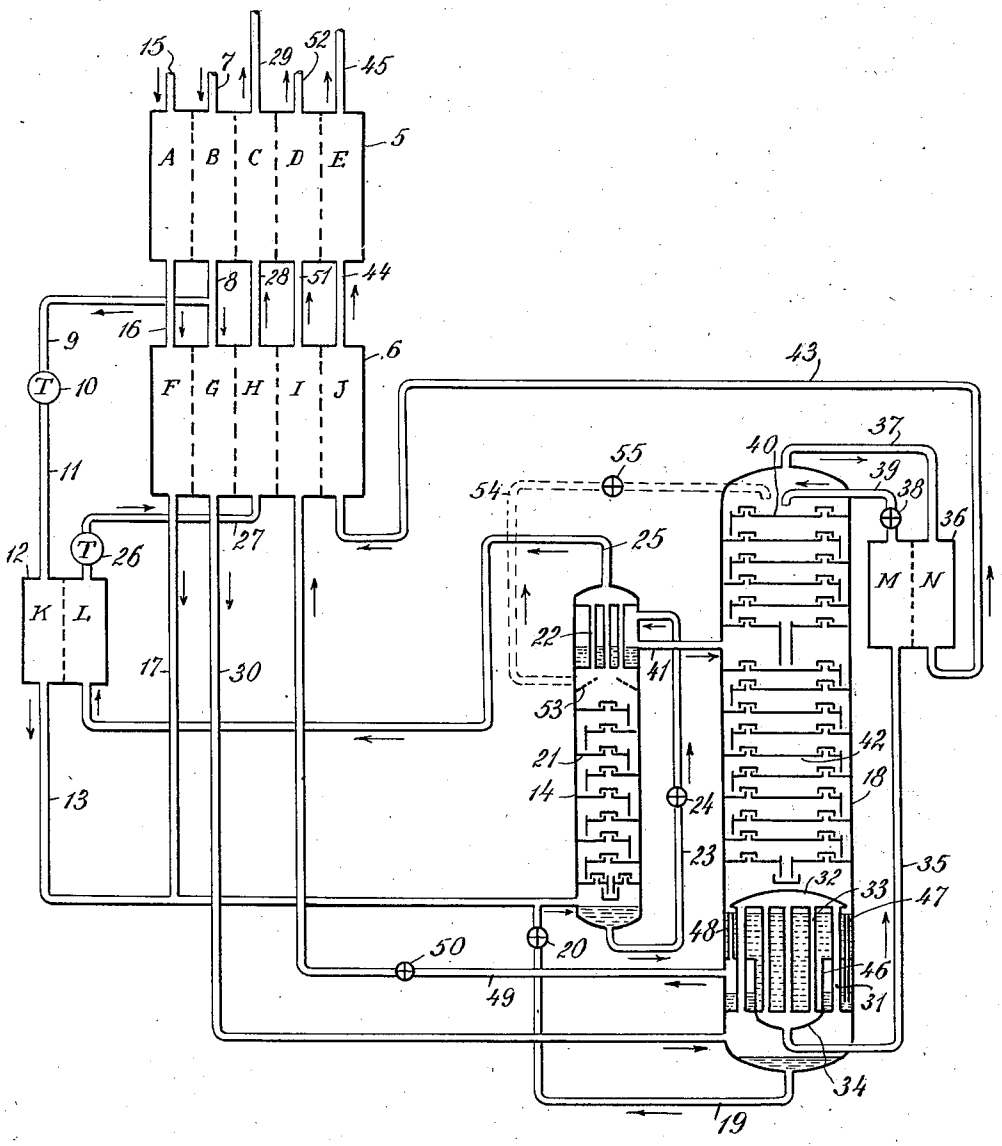

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METHOD OF SEPARATING THE CONSTITUENTS OF GASEOUS MIXTURES

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This invention relates to the separation of gaseous mixtures by liquefaction and rectification, and particularly to a method of treating air to recover the oxygen in an economical manner.

The liquefaction method of separating the constituents of air has been long established, but the commercial methods are subject to a number of defects and inefficiencies. These defects and inefficiencies generally express themselves by requiring a relatively large amount of power to separate air into its constituents.

It is the object of the present invention to provide a method which is efficient and economical whereby a high recovery of the oxygen in the air treated is obtained in a simple manner and with the minimum cost of compression. At the same time, a method is provided which permits, if desired, the recovery of part of the nitrogen in any commercial degree of purity higher than the nitrogen product of any ordinary liquefaction system.

Another object of the invention is the provision of a method of separating the constituents of gaseous mixtures wherein numerous inefficiencies of the existing systems are avoided.

Another object of the invention is to recover simultaneously with the oxygen a large fraction of the nitrogen in the air treated, in a high degree of purity, with only a small amount of added expense.

Other objects and advantages of the invention will be apparent as it is better understood by reference to the following specification and the accompanying drawing, which illustrates diagrammatically the apparatus which may be utilized in the practice of the method described. It will be understood that the drawing is merely illustrative and that details well known in the art have been omitted for the purpose of clarity.

Referring to the drawing, an exchanger 5 is represented as comprising compartments A, B, C, D and E, and a similar exchanger 6 is similarly represented as comprising compartments F, G, H, I and J. The conventional arrangement can be used whereby heat exchange is effected between the gases entering and leaving the apparatus. The entering gaseous mixture, for example air, is introduced in two parts. One part, initially compressed to a pressure of for example 5 atmospheres absolute and cooled, is introduced through pipe 7 to the compartment B and passes thence through a pipe 8 and branch 9 to an expansion turbine 10 where the gaseous mixture is expanded to a pressure of for example 2 atmospheres absolute and thereby cooled. A pipe 11 conveys the gaseous mixture to an exchanger 12 having compartments K and L, and thence through a pipe 13 to the bottom of an auxiliary rectification column 14. Another portion of the air, compressed to a pressure of for example 2 atmospheres absolute and cooled, enters through a pipe 15, passes through the compartment A of exchanger 5, thence through a pipe 16 to the compartment F of the exchanger 6, and is delivered by a pipe 17 to the pipe 13 where it joins the air which has been expanded in the turbine 10 and is delivered to the bottom of the auxiliary rectifier 14.

A portion of the air entering the bottom of the auxiliary rectifier 14 may be in the liquid phase, and the liquid is augmented by oxygen-enriched liquid delivered from the main rectifier 18 through a pipe 19 and valve 20. The balance of the air passes upwardly through the auxiliary rectifier 14 and trays 21 thereof in contact with descending liquid produced as hereinafter described. The rectification effected results in the accumulation of more liquid in the bottom of the rectifier in which liquid is enriched in oxygen, and a vapor principally nitrogen, which passes up through condenser tubes 22 at the top of the auxiliary rectifier 14. The tubes are partially surrounded by liquid supplied from the bottom of the rectifier through a pipe 23 and valve 24. The cooling of the vapor results in the provision of a liquid containing nitrogen and oxygen and any other constituents of the gaseous mixture which flows downwardly over the trays 21 to effect the rectification already described. The effluent, consisting of nitrogen of substantial purity, i. e, upward to 99.9%, escapes through a pipe 25 and is delivered to the compartment L of the exchanger 12. This nitrogen effluent is still at the pressure of the auxiliary rectifier and is expanded in a turbine 26 and thereby cooled. It passes thence through a pipe 27 to the compartment H of the exchanger 6, and through a pipe 28 to the compartment C of the exchanger 5. In the exchangers, the nitrogen serves to cool the incoming air. It is withdrawn through a pipe 29 and conveyed to any suitable storage compartment for use as may be required. Thus, the method affords immediately a quantity of high purity nitrogen as one of the products of the separation.

Another portion of the air at relatively high pressure is delivered to the compartment G of the exchanger 6, and thence through a pipe 30 to the bottom of the rectifier 18. It passes upwardly through tubes 31 of a condenser, the tubes

being surrounded by cold liquid produced by the rectification, and a portion of the air is liquefied to produce an enriched oxygen mixture which accumulates in the bottom of the rectifier and is withdrawn through the pipe 19 as hereinbefore described. The unliquefied portion of the air passes into a head 32 and downwardly through tubes 33 which are also surrounded by cold liquid products, with resulting liquefaction of the balance of the air which consists principally of nitrogen. The liquid nitrogen collects in a bowl 34 from which it is withdrawn through a pipe 35 and delivered to the compartment M of an exchanger 36, wherein it is subjected to cooling by the gaseous effluent from the column escaping through a tube 37 and passing through the compartment N of the exchanger 36. The cold liquid is delivered through a valve 38 and pipe 39 to the top of the rectifier 18 and serves as the reflux liquid for the rectification.

The liquid flows downwardly over the trays 40 within the rectifier and is augmented by liquid delivered through a pipe 41 from the condenser at the top of the auxiliary rectifier 14. The combined liquids flow downwardly over trays 42. The rectification results in the gradual enrichment of the liquid in oxygen and the accumulation of the nitrogen, a small portion of oxygen and other constituents of the atmosphere in the effluent which escapes through the pipe 37. The effluent, after passing through the compartment N of the exchanger 36, is delivered by a pipe 43 to the compartment J of the exchanger 6, and thence through a pipe 44 to the compartment E of the exchanger 5. It is withdrawn through a pipe 45 and delivered to the atmosphere as waste nitrogen.

The liquid which descends in the rectifier 18 accumulates in a receptacle 46 and overflows through a pipe 47 into the surrounding space. Some of the vapor from this liquid escapes through a pipe 48 and rises through the trays of the rectifier. The remainder is withdrawn through a pipe 49 and valve 50 and is delivered to the compartment I of the exchanger 6. It passes through a pipe 51 to the compartment D of exchanger 5 and is withdrawn through a pipe 52 and delivered to any suitable storage receptacle. This vapor constitutes the oxygen product of the method. If it is desired to supply more reflux nitrogen liquid at the top of the rectifier 18, a collector 53 indicated in dotted lines may be disposed in the auxiliary rectifier 14 to collect some of the nitrogen liquid descending from the tubes 22. This liquid may be delivered through a pipe 54, likewise indicated in dotted lines, and a valve 55, to the top of the rectifier 18.

From the above detailed description of the process it is seen that one portion of the air undergoes an initial separation, a feature common to many efficient air separation systems, into two liquid fractions, one containing substantially all of the oxygen in this portion of the air, the other being substantially pure nitrogen. The latter liquid fraction is used as reflux to the main column in the ordinary manner. The enriched oxygen liquid, however, is throttled to an intermediate pressure with the production of some vapor, and the other portion of the air treated is added thereto. The mixed vapor is subjected to a separate rectification wherein an effluent is produced which may be as pure nitrogen as is commercially demanded, and an oxygen enriched liquid fraction. The reflux for this auxiliary rectification is produced in a dephlegmator by evaporation of a part of the oxygen enriched liquid throttled to the pressure prevailing outside the dephlegmator tubes. The liquid and vapor mixture leaving the outside of the dephlegmator tubes is introduced to the main rectifier, operating at the lowest pressure in the rectification system, where it is then rectified in the usual manner by the reflux resulting from the initial separation of a part of the air.

What has been accomplished by this procedure is that there has been introduced into the middle of the main rectifier a combined liquid and vapor stream containing substantially all of the oxygen in the treated air but considerably smaller in amount and much higher in oxygen composition than is ordinarily done. Therefore, less reflux is required at the top of the main rectifier and at the same time there is available from the top of the tubes of the dephlegmator a large portion of the nitrogen entering the system at a pressure considerably higher than exists in the main rectifier. This nitrogen under pressure is available for the production of refrigeration. The net result is a considerable decrease in the work of compression required to maintain the necessary refrigeration in the system, and at the same time high recovery of the oxygen in all of the air treated is attained.

If desired, the effluent from the top of the auxiliary rectifier may be made to contain very large percentages of nitrogen, ranging upward to 99.8 to 99.9% at slight additional cost. This process thus permits oxygen to be produced in commercial purity with a simple apparatus and low compression cost, while at the same time a large fraction of the nitrogen in the air treated may be recovered, if desired, in a high degree of purity.

Various changes may be made in the procedure and particularly in the apparatus employed therefor, without departing from the invention or sacrificing any of the advantages thereof.

I claim:

1. The method of separating nitrogen and oxygen from the atmosphere by liquefaction and rectification which comprises compressing and cooling a portion of the air at relatively low pressure, compressing and cooling another portion at a relatively higher pressure, expanding a part of the air at relatively higher pressure, adding the expanded air to the portion at lower pressure, rectifying the portion at the lower pressure with liquefied constituents thereof to separate the major portion of the nitrogen as a gaseous effluent of substantial purity, separating the portion at the relatively higher pressure into liquid fractions, rectifying such fractions after expansion with the liquid product of the first mentioned rectification, discarding the gaseous effluent containing nitrogen, oxygen and other constituents of the atmosphere and evaporating the liquid product to provide oxygen of substantial purity.

2. In the method of separating air into its main constituents, oxygen and nitrogen, in which the air is initially separated in a main rectifier into two liquid fractions in which oxygen and nitrogen, respectively, are concentrated; the improvement which comprises rectifying, in an auxiliary rectifier, a substantial additional quantity of gaseous unseparated air with a portion of the top product of said rectification liquefied by thermal contact with the oxygen-enriched product of the auxiliary rectifier mixed with the oxygen-enriched fraction resulting from the initial

separation in the main rectifier, and admitting to an intermediate level of the main rectifier the oxygen-enriched mixed vapor and liquid resulting from said thermal contact in said auxiliary rectifier.

3. In the method of separating air into its main constituents, oxygen and nitrogen, in which the air is initially separated in a main rectifier into two liquid fractions in which oxygen and nitrogen, respectively, are concentrated; the improvement which comprises rectifying, in an auxiliary rectifier, a substantial additional quantity of gaseous unseparated air with a portion of the top product of said rectification liquefied by thermal contact with the oxygen-enriched product of the auxiliary rectifier mixed with the oxygen-enriched fraction resulting from the initial separation in the main rectifier, withdrawing the uncondensed top product of the auxiliary rectifier as high purity nitrogen, and admitting to an intermediate level of the main rectifier the oxygen-enriched mixed vapor and liquid resulting from said thermal contact in said auxiliary rectifier.

4. In the method of separating air into its main constituents, oxygen and nitrogen, in which the air is initially separated in a main rectifier into two liquid fractions in which oxygen and nitrogen, respectively, are concentrated; the improvement which comprises rectifying, in an auxiliary rectifier, a substantial additional quantity of gaseous unseparated air with a portion of the top product of said rectification liquefied by thermal contact with the oxygen-enriched product of the auxiliary rectifier mixed with the oxygen-enriched fraction resulting from the initial separation in the main rectifier, withdrawing the uncondensed top product of said auxiliary rectifier and expanding it for refrigeration purposes, and admitting to an intermediate level of the main rectifier the oxygen-enriched mixed vapor and liquid resulting from said thermal contact in said auxiliary rectifier.

5. In the method of separating air into its main constituents, oxygen and nitrogen, in which the air is initially separated in a main rectifier into two liquid fractions in which oxygen and nitrogen, respectively, are concentrated; the improvement which comprises rectifying, in an auxiliary rectifier, a substantial additional quantity of gaseous unseparated air with a portion of the top product of said rectification liquefied by thermal contact with the oxygen-enriched product of the auxiliary rectifier mixed with the oxygen-enriched fraction resulting from the initial separation in the main rectifier, diverting a portion of the liquid produced at the top of the auxiliary rectifier to the top of the main rectifier to supplement the reflux liquid available in said main rectifier, and admitting to an intermediate level of the main rectifier the oxygen-enriched mixed vapor and liquid resulting from said thermal contact in said auxiliary rectifier.

6. In the method of separating air into its main constituents, oxygen and nitrogen, in which the air is initially separated in a main rectifier into two liquid fractions in which oxygen and nitrogen, respectively, are concentrated; the improvement which comprises expanding a substantial additional quantity of high pressure gaseous unseparated air to a lower pressure, rectifying said expanded air in an auxiliary rectifier with a portion of the top product of said rectification liquefied by thermal contact with the oxygen-enriched product of the auxiliary rectifier mixed with the oxygen-enriched fraction resulting from the initial separation in the main rectifier, and admitting to an intermediate level of the main rectifier the oxygen-enriched mixed vapor and liquid resulting from said thermal contact in said auxiliary rectifier.

7. In the method of separating air into its main constituents, oxygen and nitrogen, in which the air is initially separated in a main rectifier into two liquid fractions in which oxygen and nitrogen respectively are concentrated; the improvement which comprises rectifying, in an auxiliary rectifier, a substantial additional quantity of unseparated air with a portion of the top product of said rectification liquefied by thermal contact with the oxygen enriched fraction resulting from the initial rectification in the main rectifier, and admitting to an intermediate level of the main rectifier the oxygen enriched mixed vapor and liquid resulting from said thermal contact in said auxiliary rectifier.

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