

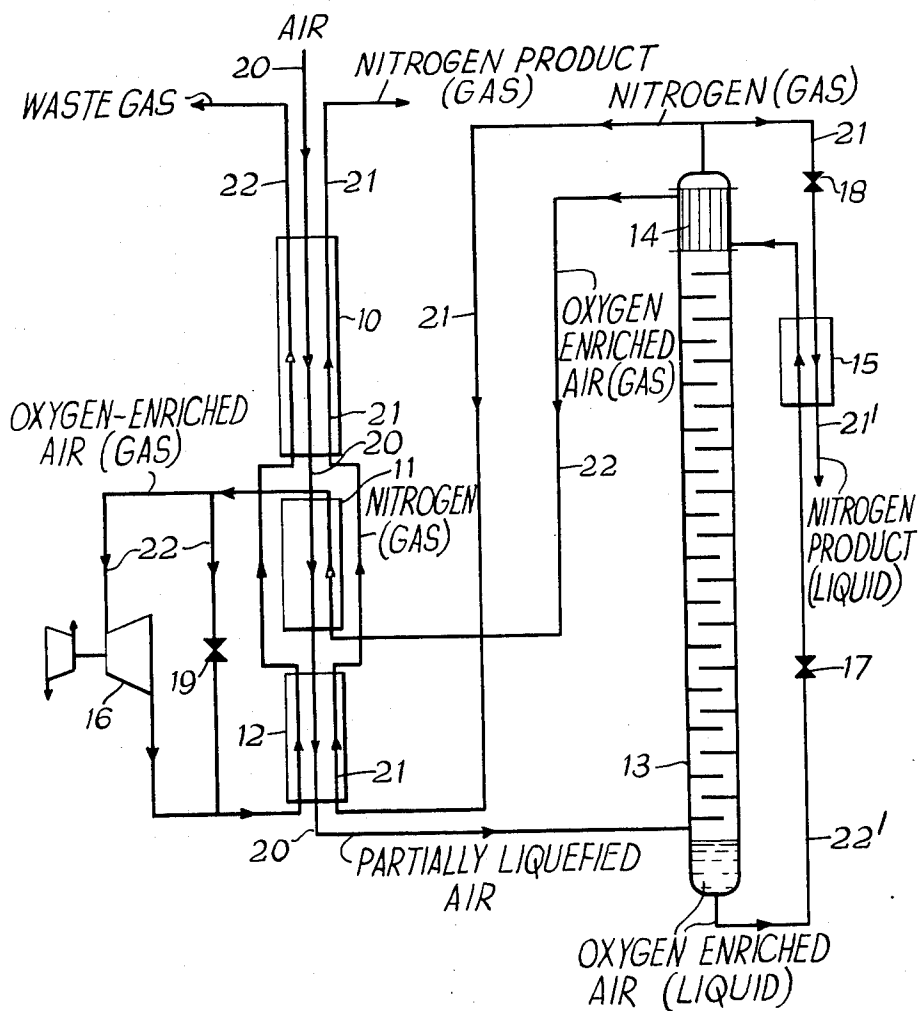
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PRODUCTION OF NITROGEN

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PRODUCTION OF NITROGEN

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This invention is concerned with the production of nitrogen from air by low temperature methods of separation.

There is now an increasing demand from many industries for moderate quantities of pure nitrogen, to be used for blanketing vessels, purging and other purposes. The requirements are large enough to render uneconomical the transport from a distance of compressed gas in bottles or of liquid nitrogen in tanks; on the other hand, the conventional methods of producing nitrogen on site are not always satisfactory.

Plants based on the combustion of fuels and the subsequent purification of the flue gases show a high cost of utilities and it is difficult, with such plants, to guarantee a purity of 99.9% and over.

The known methods of producing nitrogen by low temperature separation of air whether using a single fractionation column or a double fractionation column have various disadvantages. Thus they normally require a high initial compression of the air to be separated, requiring the use of multi-stage compressors and reciprocating expanders. Most of the known methods also yield the nitrogen at atmospheric pressure, so that if the nitrogen has to be recompressed for storage or for use, additional compressors have to be installed and, unless special precautions are taken, the initially pure product may easily be contaminated. Furthermore the methods hitherto suggested for producing the cold required for operation of the low temperature processes are such as to make it extremely difficult to vary the yield in accordance with a fluctuating demand or to keep the plant running without production of nitrogen when there is a temporary cessation of demand.

It is an object of this invention to provide a method whereby nitrogen of a high degree of purity may be produced at a substantial superatmospheric pressure and in an economical manner by low temperature separation of air.

It is a further object of the invention to provide such a method whereby both gaseous and liquid nitrogen may be produced.

It is still a further object of the invention to provide such a method which can easily and simply be controlled to vary the output of nitrogen in accordance with a fluctuating demand.

The present invention provides an improved and highly flexible method of producing pure nitrogen at a substantial superatmospheric pressure by the low temperature separation of air. A single fractionating column is employed and the air is initially compressed to a pressure which is substantially the same as that at which the column operates, the difference between the latter pressure and the initial pressure being solely due to pressure losses in the system. The pressure may be a relatively low one, such as 8 atmospheres, which can be obtained with a two-stage compressor. The air is freed from CO₂ and moisture by conventional means, such as scrubbing and cooling, before it enters the low temperature system.

The important feature of the method of the invention is the manner in which the cold required for the separation is produced without additional utilities consumption. This is achieved by separating the air in the fractionating column into highly pure nitrogen gas as overhead product and oxygen-enriched liquid air as bottoms product,

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withdrawing and expanding the latter to an intermediate pressure and then evaporating it in indirect heat exchange with gas in the top of the column, whereby reflux is provided, warming up the evaporated oxygen-enriched air in partially cooling the incoming air and then expanding at least part thereof in an expansion turbine to produce cold for use in the process, the expanded, cold, oxygen-enriched air and the cold gaseous nitrogen product being used to cool down the incoming air to the required temperature.

By operating in this fashion, the output of nitrogen can be varied simply by controlling the amount of air delivered by the compressor. Furthermore the plant can be kept operating with total reflux in the column, so that no nitrogen is taken off, by control of the air supply.

In carrying out the method of the invention the following procedure may be adopted.

Air at a superatmospheric pressure, for example about 8 atmospheres, from which CO₂ and moisture have been removed by conventional methods, is passed through at least three cooling zones in series, in the last of which it is cooled to a temperature at which a minor part thereof is liquefied; the resulting gas-liquid air mixture is then fed to the base of a fractionating column operating at said superatmospheric pressure and having a condenser in the head thereof, in which the major portion of the air is liquefied as it passes up the column and flows back down the column as reflux, yielding highly pure nitrogen gas as overhead product and leaving oxygen-enriched liquid air as bottoms product; the liquid bottoms product is withdrawn from the base of the fractionating column and is expanded to a pressure intermediate atmospheric pressure and said superatmospheric pressure and then passed through the condenser of the fractionating column in indirect heat exchange contact with nitrogen in the head of the column whereby part of the nitrogen is condensed and the oxygen-enriched liquid air is evaporated; the evaporated oxygen-enriched air is then passed in indirect heat exchange contact with the incoming air through an intermediate zone of said cooling zones, in which it is warmed up, giving up cold, and thence through an expansion turbine in which it is expanded to a lower pressure and thereby cooled; the finally expanded cold oxygen-enriched air is then passed in indirect heat exchange contact with the incoming air first through the final zone of said cooling zones in which the air is partially liquefied and then through at least the first of said cooling zones; the highly pure nitrogen gas leaving the top of the fractionating column at superatmospheric pressure is passed in indirect heat exchange contact with the incoming air through at least the first of said cooling zones and then collected as a gaseous product under superatmospheric pressure.

In a modification of this process designed to produce a small proportion, for example up to 10% of the nitrogen product as liquid, part of the nitrogen gas stream leaving the top of the fractionating column is directed to pass in indirect heat exchange with the expanded oxygen-enriched liquid air before the latter is passed into the condenser of the fractionating column. Part of the oxygen-enriched liquid air is evaporated in condensing the diverted nitrogen stream which is then collected as a liquid product under superatmospheric pressure.

Other features of the invention will become apparent from the following detailed description made with reference to the accompanying drawing and illustrating, by way of example, the manner in which the invention may be carried out.

The accompanying drawing illustrates schematically one form of apparatus in accordance with the invention.

Referring to the drawing, 10, 11 and 12 are three heat exchangers through which the incoming air is passed in series, 13 is a fractionating column provided with a condenser 14 at the head thereof, the base of the fractionating column being connected to the air outlet of heat exchanger 12. 15 is an additional heat exchanger, 16 is an expansion turbine, 17 and 19 are expansion valves and 18 is a valve.

The path of the input air is represented by the line 20, that of the nitrogen gas product by the line 21, the liquid nitrogen product being represented by 21'. The path of the oxygen-enriched air or residual waste gas is represented by the line 22 when in gaseous form and by the line 22' when in liquid form.

The feed or input air is at a pressure of 8 atmospheres absolute and has been freed from CO₂ and dried. Compression to 8 atmospheres absolute may be carried out in conventional manner in a two-stage reciprocating compressor and carbon dioxide may be removed by scrubbing in known manner with caustic-soda solution. Drying may also take place in conventional manner by means of alumina driers, with precooling, if desired, to about 2 to 5° C., to first condense out the major portion of the water vapour. A separate refrigerating unit of known type e.g. a Freon unit, may be used for this precooling. The compressing, scrubbing and drying means are conventional and are accordingly not illustrated.

The air 20, freed from CO₂ and moisture, at 8 atmospheres absolute pressure and at ambient temperature or at a temperature of about 2 to 5° C., is passed through the heat exchangers 10, 11 and 12 in turn. It is cooled to about 120° K. (-153° C.) in exchanger 10, to about 114° K. (-159° C.) in exchanger 11 and to about 105° K. (-168° C.) in exchanger 12, in which a minor portion of the air is liquefied. The partially liquefied air 2 leaving heat exchanger 12 is fed to the base of fractionating column 13 operating at about 8 atmospheres absolute, in which the major portion of the rising air is liquefied and flows down the column as reflux yielding highly pure nitrogen gas as overhead product. The bottom product which consists of oxygen-enriched liquid air, containing, for example, about 31% oxygen, is withdrawn through line 22' and expanded through expansion valve 17 to about 5 atmospheres absolute and then passed via heat exchanger 15 to condenser 14 in which it is evaporated in condensing part of the nitrogen gas in the head of column 13. The evaporated oxygen-enriched air or waste gas 22 is passed through heat exchanger 11, in which it is warmed up in cooling the incoming air and then through expansion turbine 16 in which it is expanded to about 1.3 atmospheres absolute and thereby cooled and the resulting cold gas is passed through heat exchanger 12, in which it is warmed up, in cooling and partially liquefying the incoming air, and thence through heat exchanger 10, in which it is further warmed up. Additional cold is provided in heat exchangers 10 and 12 by the nitrogen gas stream leaving the top of column 13, which is passed as shown by line 21 through these exchangers and is then collected at a pressure of about 8 atmospheres absolute.

When the plant is functioning to produce only gaseous nitrogen the valve 18 is closed and all the nitrogen gas from the top of fractionating column 13 is passed to heat exchanger 12. In this case only part, though a major part, of the evaporated oxygen-enriched air 22 passes through the turbine 16, the remainder being by-passed through expansion valve 19, the pressure at the inlet to the turbine being correspondingly reduced. The minor part of the gas passing through expansion valve 19 is expanded to the same pressure as the major part which passes through the turbine 16. When part of the nitrogen product is to be produced as liquid, about 10% of the nitrogen gas leaving the top of column 13 may be diverted through valve 18 to be cooled and condensed in heat exchanger 15. The remainder of the nitrogen

gas is passed to heat exchanger 12 and thence via exchanger 10 to the outlet. For this mode of operation all the evaporated oxygen-enriched gas leaving the heat exchanger 11 is expanded through the turbine 16.

About one third of the air is delivered as substantially pure nitrogen at about 8 atmospheres absolute pressure at the top of the fractionating column, the remaining two thirds being available for the production of cold.

The gaseous nitrogen product is passed to a suitable pressure storage vessel from which it may be withdrawn for use. Production of nitrogen may take place simultaneously with withdrawal for use, in which case pressure control means may be used to control the operation of the plant in accordance with the take-off of nitrogen gas for use.

The production of part of the nitrogen product in liquid form is useful in that it enables a store of liquid nitrogen to be accumulated against a shut-down of the plant.

Though starting up of the plant may take place with one turbine as illustrated, an additional turbine may be provided operating in parallel with turbine 16 to hasten the attainment of the required operating conditions.

All the heat exchangers used in the plant including the reflux condenser 14 are of the indirect heat exchange type and may suitably be of the corrugated-fin type, made of aluminium, as supplied by Marston Excelsior Limited of Wolverhampton, England. The fractionating column itself may have a stainless steel shell on account of the relatively high operating pressure and be fitted with conventional, closely spaced, brass sieve plates. The cold box housing the parts illustrated is suitably lagged, for example, with Brilite.

The expansion turbine is suitably of the radial inward flow type with an optimum running speed of the order of 40,000 to 45,000 r.p.m. Such turbines suitable for operating at extremely low temperatures and performing the duties required are manufactured by Joseph Lucas (Hydraulic and Combustion Equipment) Limited of Birmingham, England.

The process and plant described above give a highly pure nitrogen product at an elevated pressure with a very low utilities consumption, the refrigeration required for the operation of the process being supplied by the process itself.

We claim:

1. A process for producing highly pure nitrogen at a substantial superatmospheric pressure from air which has been compressed to about said superatmospheric pressure and from which CO₂ and moisture have been removed, comprising the steps of:

- (a) cooling said air at about said superatmospheric pressure to a temperature at which a minor part of said air is liquefied;
- (b) supplying the resulting partially liquefied air to the base of a single fractionating column having a condenser for providing reflux at the upper end thereof and operating at about said superatmospheric pressure;
- (c) separating said supplied air in said column into a highly pure nitrogen gas as overhead product and an oxygen-enriched liquid air as bottoms product;
- (d) withdrawing said oxygen-enriched liquid air from the bottom of said column and expanding the same to a pressure intermediate said superatmospheric pressure and atmospheric pressure;
- (e) passing said expanded oxygen-enriched liquid air through said condenser in indirect heat exchange with the nitrogen gas separated in the upper end of said column whereby said oxygen-enriched liquid air is evaporated in said condenser in condensing nitrogen gas to provide reflux for said column;
- (f) withdrawing said evaporated oxygen-enriched liquid air from said condenser and passing the same in indirect heat exchange with incoming air to cool

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said incoming air and warm said evaporated oxygen-enriched liquid air;

(g) then expanding at least a part of said warmed oxygen-enriched air to a pressure close to atmospheric pressure in an expansion turbine thereby to produce cold for use in said process;

(h) utilizing the oxygen-enriched air at close to atmospheric pressure to cool incoming air by indirect heat exchange and then venting it from the process;

(i) withdrawing gaseous nitrogen product from the top of said column;

(j) passing at least a part of said withdrawn gaseous nitrogen product in indirect heat exchange with incoming air to cool said incoming air; and

(k) then collecting at least said part of said withdrawn gaseous nitrogen product at substantially said superatmospheric pressure.

2. A process in accordance with claim 1 wherein the superatmospheric pressure is at least about eight atmospheres absolute.

3. A process in accordance with claim 1 wherein the air is introduced to the process at a reduced temperature between 0° C. and ambient temperature.

4. A process in accordance with claim 1 wherein a major portion of the evaporated oxygen-enriched air is passed through the expansion turbine and a minor portion of said oxygen-enriched air is passed through an expansion valve and the resulting expanded gases are recombined prior to passing said expanded gas in indirect heat exchange with the incoming air, as set forth in step (h).

5. A process in accordance with claim 1 wherein a major portion of the evaporated oxygen-enriched air is passed through the expansion turbine and a minor portion of said oxygen-enriched air is passed through an expansion

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valve, the resulting expanded gases are recombined prior to passing said expanded gases in indirect heat exchange with the incoming air, as specified in step (h) and only gaseous nitrogen product is recovered.

6. The process in accordance with claim 1 wherein a minor portion of the nitrogen product leaving the fractionating column is liquefied and collected as a liquid product.

7. A process in accordance with claim 1 wherein a minor portion of the nitrogen product leaving the fractionating column is passed in indirect heat exchange with oxygen-enriched liquid air from the bottom of said column after it has been expanded in accordance with step (d).

8. A process in accordance with claim 1 wherein a minor part of the nitrogen product leaving the top of the fractionating column is passed in indirect heat exchange with oxygen-enriched liquid air from the bottom of said column after it has been expanded in accordance with step (d) and all of the evaporated oxygen-enriched air is passed through the expansion turbine.

References Cited by the Examiner

UNITED STATES PATENTS

1,594,336	7/26	Newes	62—31
2,040,107	5/36	Schlitt	62—39 X
2,520,862	8/50	Swearingen	62—38 X
2,626,510	1/53	Schilling	62—38 X
2,688,238	9/54	Schilling	62—31 X
2,850,880	9/58	Jakob	62—39 X
3,070,966	1/63	Ruhemann	62—39 X

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