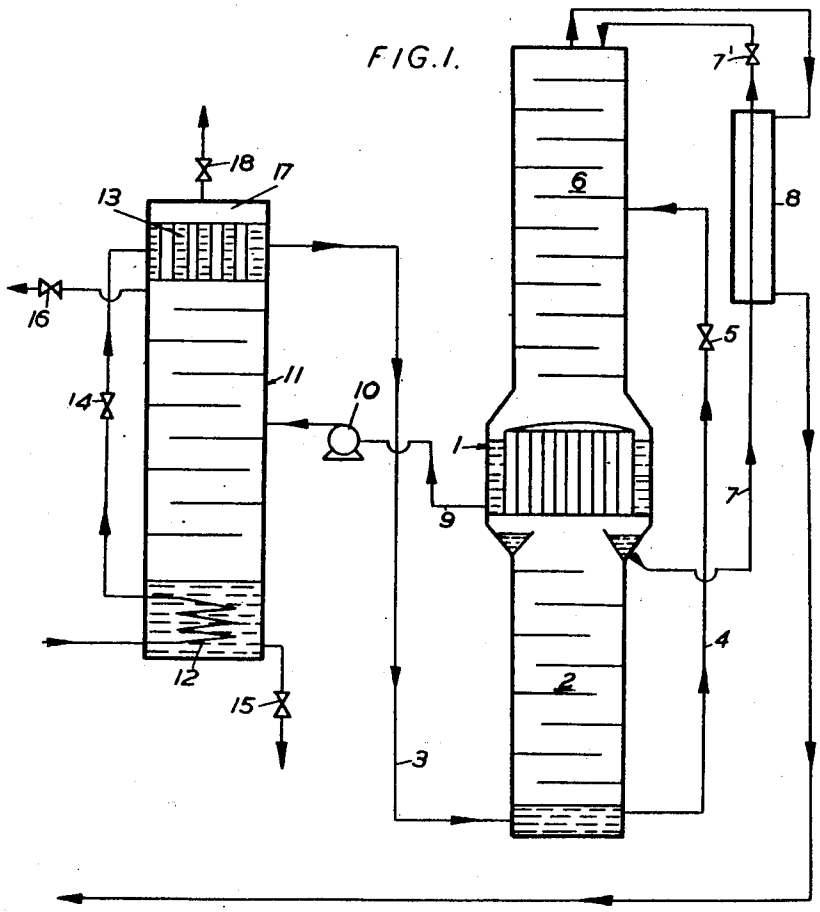
July 3, 1951 N. W. ROBERTS 2,559,132

FRACTIONAL SEPARATION OF AIR

Filed Feb. 8, 1949 2 Sheets-Sheet 1

FIG. 1.



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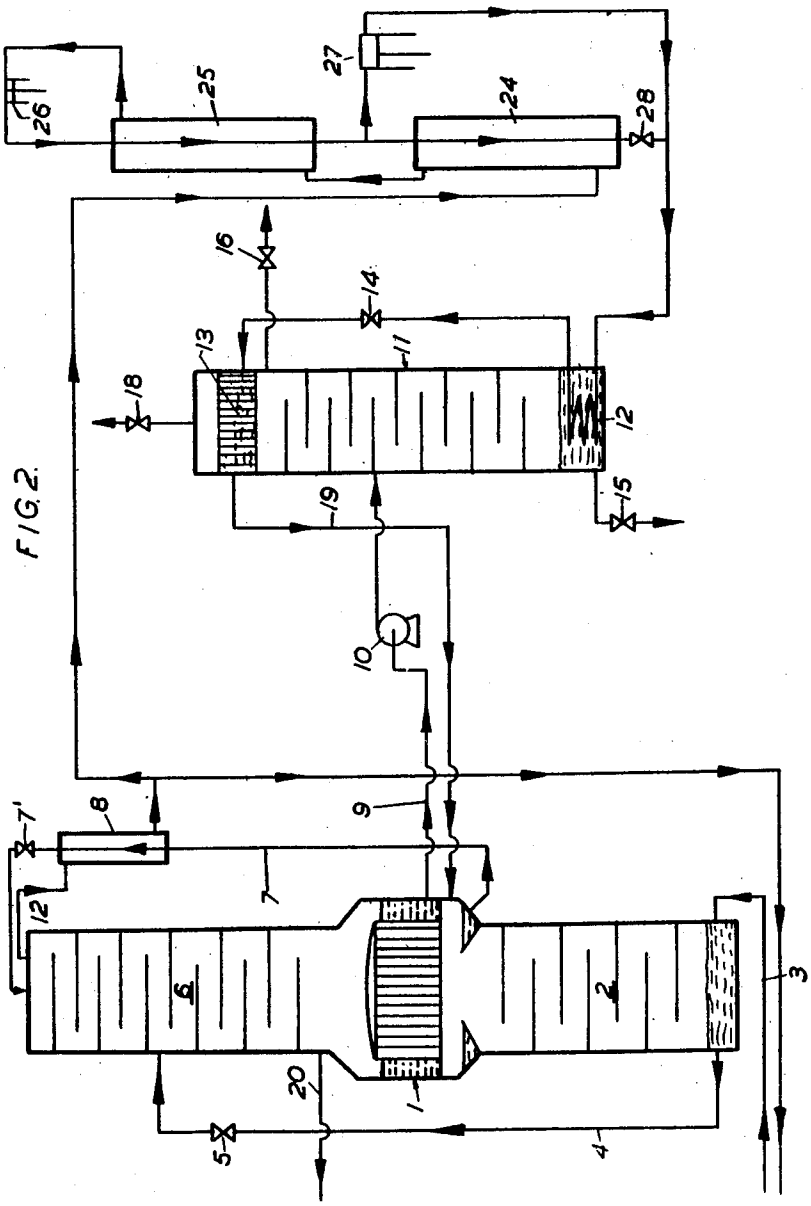


FIG. 2.

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FRACTIONAL SEPARATION OF AIR

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1 Claim. (Cl. 62—175.5)

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The present invention relates to the fractional separation of air.

In the fractional separation of air there is frequently employed a rectification process by means of which three fractions respectively rich in nitrogen, argon and oxygen are obtained. Such a separation is usually carried out in the well-known double column in which the liquid nitrogen fraction separated in the lower column is after expansion introduced into the upper column to serve as reflux. For the production of the argon fraction, however, it is necessary to add to the double column an auxiliary column, the reflux for which is obtained by indirect heat exchange with either a part of the liquid nitrogen fraction or with a part of the so-called "rich liquid" obtained from the lower column.

In such a double column separation process where the reflux medium is divided between the upper column and the aforesaid auxiliary column, there is not sufficient reflux available to make possible the separation of all three constituents in high yield and at a high purity. The separation of a fraction rich in argon in an auxiliary column as described above requires reflux which can only be obtained by diminishing the amount of reflux available for the double column separation and consequently by decreasing the yield or purity of either the oxygen-rich or nitrogen-rich fraction.

The complete separation of the air into its three main components can only be effected by the expenditure of more power than is required to operate the aforesaid double column separation process.

According to this invention there is provided a process for separating argon from air which comprises treating the air in a primary rectification zone in such a manner as to produce therein as one of the separation products a liquid argon-oxygen fraction, increasing the pressure of such fraction and transferring it to a secondary rectification zone which operates under elevated pressure wherein it is separated into an argon-rich fraction and a substantially pure oxygen fraction, the necessary temperature gradient in the said secondary rectification zone being maintained by the indirect condensation and revaporisation of a fluid, the revaporisation taking place at such a pressure that the vaporised fluid can act as reflux medium in the primary rectification zone.

The fluid may be one of the separation products obtained in the primary rectification zone or it may be air. It will be understood that the

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fluid prior to its introduction into the secondary rectification zone must be cooled substantially to its condensation temperature.

In the specific case in which air is used as the fluid and in which the primary rectification zone operates with two-stage rectification, the air after having served to maintain the necessary temperature gradient in the secondary rectification zone may be introduced into the first stage of the two-stage rectification constituting the primary rectification zone.

Where nitrogen is used as the fluid and where the primary rectification zone operates with two-stage rectification, the nitrogen after having served to maintain the necessary temperature gradient in the secondary rectification zone may be introduced into the first stage of the two-stage rectification constituting the primary rectification zone.

In order to secure the condensation of the air or nitrogen used as reflux medium by indirect heat exchange with the liquid argon/oxygen fraction at the base of the primary rectification zone, a minimum pressure of about 5.5 atma. is required in practice. In order to carry out the invention and to use the reflux medium evaporating in the condenser of the secondary rectification zone as reflux medium in the primary rectification zone, it is therefore necessary to evaporate said reflux medium at a pressure not less than 5.5 atma. This corresponds to a temperature at which argon vapour cannot condense unless its pressure exceeds 2 atma., but in practice somewhat more, in view of the unavoidable temperature difference for heat exchange and the traces of nitrogen present in the condenser. Accordingly it is necessary, in order to carry out the invention, to increase the pressure of the liquid argon/oxygen feed to at least this value.

A fraction rich in argon can also be recovered according to this invention where single stage rectification is carried out in the primary rectification zone, a circulating stream of nitrogen being used as reflux medium. In this case, nitrogen is the fluid which is used for maintaining the necessary temperature gradient in the secondary rectification zone and after performing this function it is introduced as reflux medium into the primary rectification zone. In all cases, the common feature is the production in a primary rectification zone of an argon-oxygen liquid fraction, the pressure of which is then increased before it is introduced into a secondary rectification zone which then operates at a pressure higher than that under which the argon-oxygen

rich fraction was separated in the primary zone.

It is known that the amount of reflux necessary to separate a given quantity of air into an oxygen-argon mixture and nitrogen is substantially less than that required to separate the oxygen-argon mixture so obtained into oxygen and argon. Thus, in the particular case in which all the oxygen withdrawn as the oxygen-argon fraction from the primary rectification zone is separated and withdrawn as pure oxygen in the secondary rectification zone, only a part of the fluid which has been used for maintaining the necessary temperature gradient in the secondary rectification zone will be needed to act as a source of reflux in the primary rectification. The remainder of this fluid which will be at its vaporisation pressure will therefore be available for other purposes.

This excess can be used for the production of additional cold by isentropic expansion, the expanded gas then being passed direct into the primary rectification zone. The cold produced by expansion can be used to undercool the liquid oxygen product from the secondary rectification zone thus avoiding undesirable flash losses.

Alternatively the excess of fluid from the secondary rectification zone may be usefully employed in assisting in the compression of the argon-oxygen liquid fraction thereby economising in compression energy.

Still another method of utilising the excess fluid from the secondary rectification zone is to carry out further rectification processes such as the production of nitrogen of extra high purity or the concentration of krypton and xenon from the oxygen obtained in the secondary rectification zone.

The invention will now be more particularly described with reference to the accompanying drawings in which

Figure 1 shows in conventional diagrammatic form an air separation process for producing an argon fraction, a nitrogen fraction and substantially pure liquid oxygen, whilst

Figure 2 shows in conventional diagrammatic form an air separation process for producing an argon fraction, a nitrogen fraction and a gaseous oxygen fraction containing 96–98% oxygen.

Referring to Figure 1, a primary rectification zone is formed by a double column 1 comprising in known manner a lower column 2 to which compressed air at its dew point is fed through a pipe 3. In the lower column 2 there accumulates at the base an oxygen-rich fraction which is transferred through pipe 4 and expansion valve 5 to the upper column 6 of the primary rectification zone, and at the upper end a nitrogen-rich fraction which is transferred through pipe 7 and expansion valve 7' to the top of the upper column, passing beforehand through a heat exchanger 8 in heat exchange relationship with the effluent nitrogen from the column 6. In the upper column the air is separated into a nitrogen fraction which leaves the top of the column and is passed through the heat exchanger 8 as aforesaid, and a liquid argon-oxygen fraction which collects at the base of the upper column. This liquid fraction is withdrawn through pipe 9, compressed in a compressor 10 and then introduced into a simple column 11 constituting a secondary rectification zone. This column is heated at the base by a coil 12 to which the air to be separated is fed at its dew point and at a pressure of about 10 atma., and cooled at the top by a condenser 13 to which the air stream cooled and condensed in the coil

12 is fed after expansion in valve 14 to about 5.5 atma.

The expansion of the liquid air through valve 14 results in a drop in its temperature so that the temperature of the liquid air in condenser 13 is such as to cause the vapours rich in argon rising in column 11 to be condensed by indirect heat exchange and to flow back as reflux.

The air leaving the condenser 13 is fed through pipe 3 to the lower column 2 of the primary rectification zone as already stated. Though the air stream will be partially vaporised in traversing the condenser 13, the total heat content and pressure are so adjusted that the air on entering the lower column 2 is substantially at its dew point whereby the double column 1 can be operated in the customary manner.

The separation effected in column 11, permits of the withdrawal from the base thereof through a valve 15 of a liquid oxygen fraction of high purity, whilst a fraction rich in argon can be withdrawn from a point at the top of column 11 immediately below the condenser 13 through valve 16. Nitrogen present in the liquid argon-oxygen fraction fed to the column 11 will collect in a header 17 above the condenser 13 whence it may be vented through valve 18.

Since the upper column 6 is so operated as to concentrate the major portion of the argon-oxygen fraction in the processed air on the lower trays instead, as heretofore, in the middle part of the column, there is an extended zone in the upper part of the column for washing residual oxygen from the rising vapours and a far reaching separation of nitrogen from the higher boiling point constituents can thereby be achieved, resulting in the production of high purity nitrogen with consequent improvement in the oxygen yield.

Alternatively, by proceeding in accordance with the invention, the separation of nitrogen from higher boiling point constituents can be achieved to the same extent in a primary rectification column with a smaller number of trays than is required when proceeding in accordance with the practice hitherto.

By proceeding therefore in the manner described, argon may be recovered in exceptionally high yield, while at the same time, liquid oxygen of high yield and purity is obtained.

A process for producing gaseous oxygen of about 96–98% purity will now be described with reference to Figure 2 where a part of the total oxygen yield may be recovered as substantially pure liquid oxygen concurrently with the recovery of a high purity argon fraction.

In this case air at its dew point is fed direct to the lower column 2 through the pipe 3 and is not, as in the previous case, first used as the boiling and condensing medium for the secondary rectification zone. Otherwise the primary rectification zone operates in the same manner as that of Fig. 1 and the various parts bear the same reference numerals except that there is provision for withdrawing a gaseous oxygen fraction through pipe 20 leaving the column 6 at a point above the liquid argon-oxygen fraction which collects at the base of the column.

The necessary reflux to operate the simple column 11 constituting the secondary rectification zone is obtained from a portion of the nitrogen leaving the top of the upper column 6. This is raised to approximately atmospheric temperature by heat exchange in exchangers 24 and 25, compressed to a high pressure in a compressor

26 and returned in counter-current flow to the cold nitrogen through heat exchanger 25. One portion of the compressed nitrogen is then expanded to a pressure of about 10 atmospheres absolute in an expansion machine 27 whereby it is further cooled, and the other portion is liquefied by counter-current heat exchange with the cold nitrogen fraction in exchanger 24. After expansion in a valve 28 to 10 atmospheres absolute pressure, this liquefied portion is mixed with the exhaust from expansion engine 27 and admitted as a mixture of vapour and liquid to the coil 12 in the base of the secondary column 11, where it is condensed by indirect heat exchange with the substantially pure liquid oxygen boiling in the base thereof. The condensed liquid nitrogen is then expanded at valve 14 to a pressure substantially equal to that of the lower column 2 of the double column 1, usually at 5.5 atmospheres absolute. The expanded nitrogen is passed through the condenser 13 where it is partially vaporized by indirect heat exchange with the vapors rich in argon at the top of the secondary column 11. These argon-rich vapours are thereby condensed and returned as reflux to the secondary column. The nitrogen leaving the condenser 13 is returned to the upper end of the lower column 2 where it is totally condensed and used as reflux in the customary manner.

A substantially pure liquid oxygen product is withdrawn from the secondary column 11 through valve 15 and a fraction rich in argon is withdrawn from the top of the secondary column 11 through valve 16. Traces of nitrogen which might collect at the top of the secondary column 11 may be vented through valve 18.

It will be clear that the primary rectification zone may be a single column using circulating nitrogen as reflux medium, instead of a double column, the arrangement of the secondary rectification zone and additional refrigeration system being substantially as described with reference to Fig. 2.

It is understood that the specific arrangements described with reference to Figures 1 and 2 are only illustrative and are not to be construed so as to limit the invention to the specific embodiments described.

I claim:

A process for separating argon from air which comprises treating the air in a two-stage primary rectification zone in such a manner as to produce therein as one of the separation products a liquid oxygen-argon fraction, increasing the pressure of such fraction and transferring it to a secondary rectification zone which operates under elevated pressure wherein it is separated into an argon-rich fraction and a substantially pure oxygen fraction, maintaining the necessary temperature gradient in the said secondary rectification zone by the condensation and revaporization of air by indirect heat exchange therein, and introducing the revaporized air into the first stage of the two-stage primary rectification zone.

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