

June 5, 1934.

W. L. DE BAUFRE

1,961,202

APPARATUS FOR SEPARATING MIXED GASES BY LIQUEFACTION

Filed March 17, 1932

3 Sheets-Sheet 1

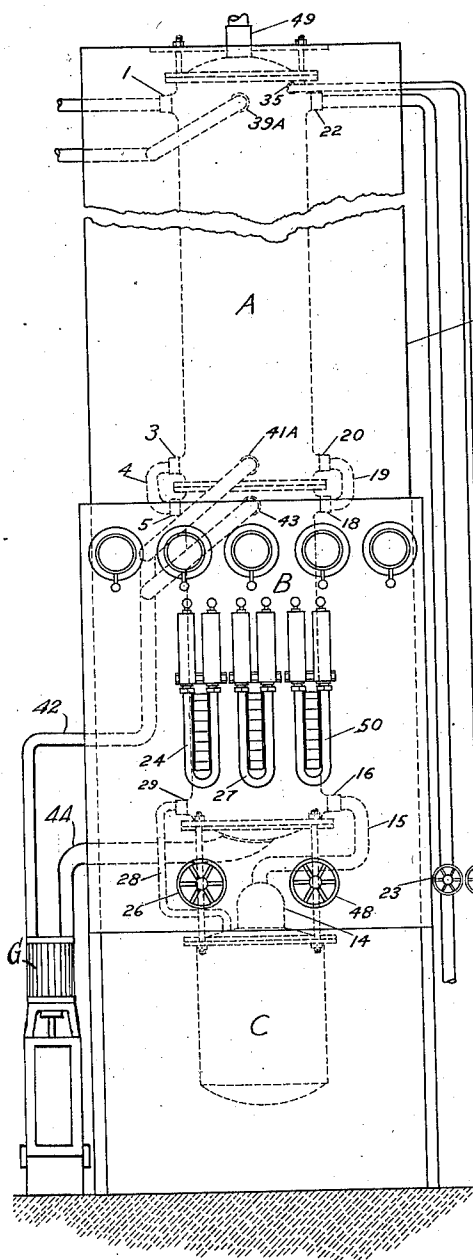
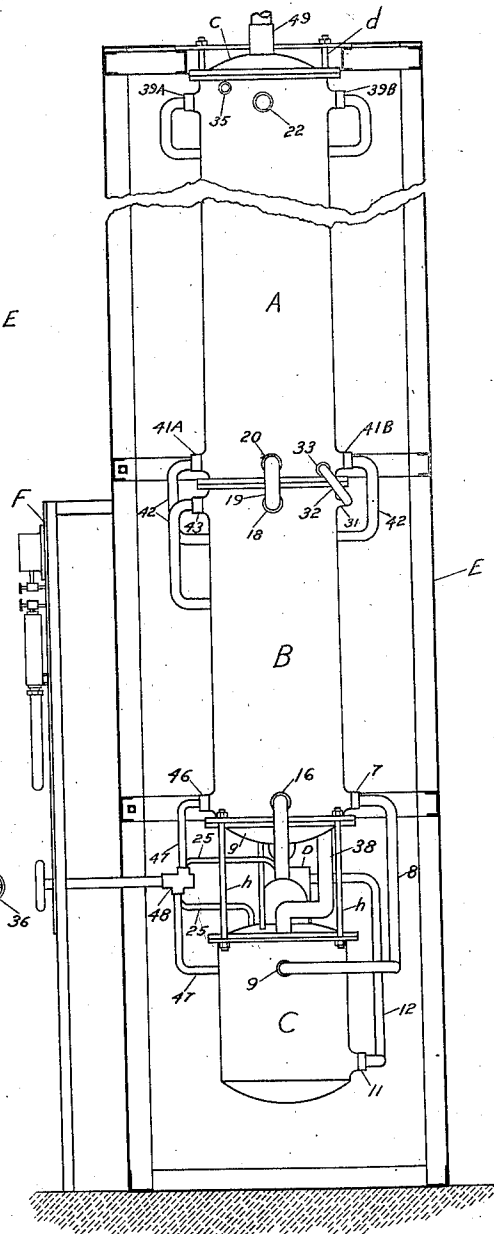


Fig. 2
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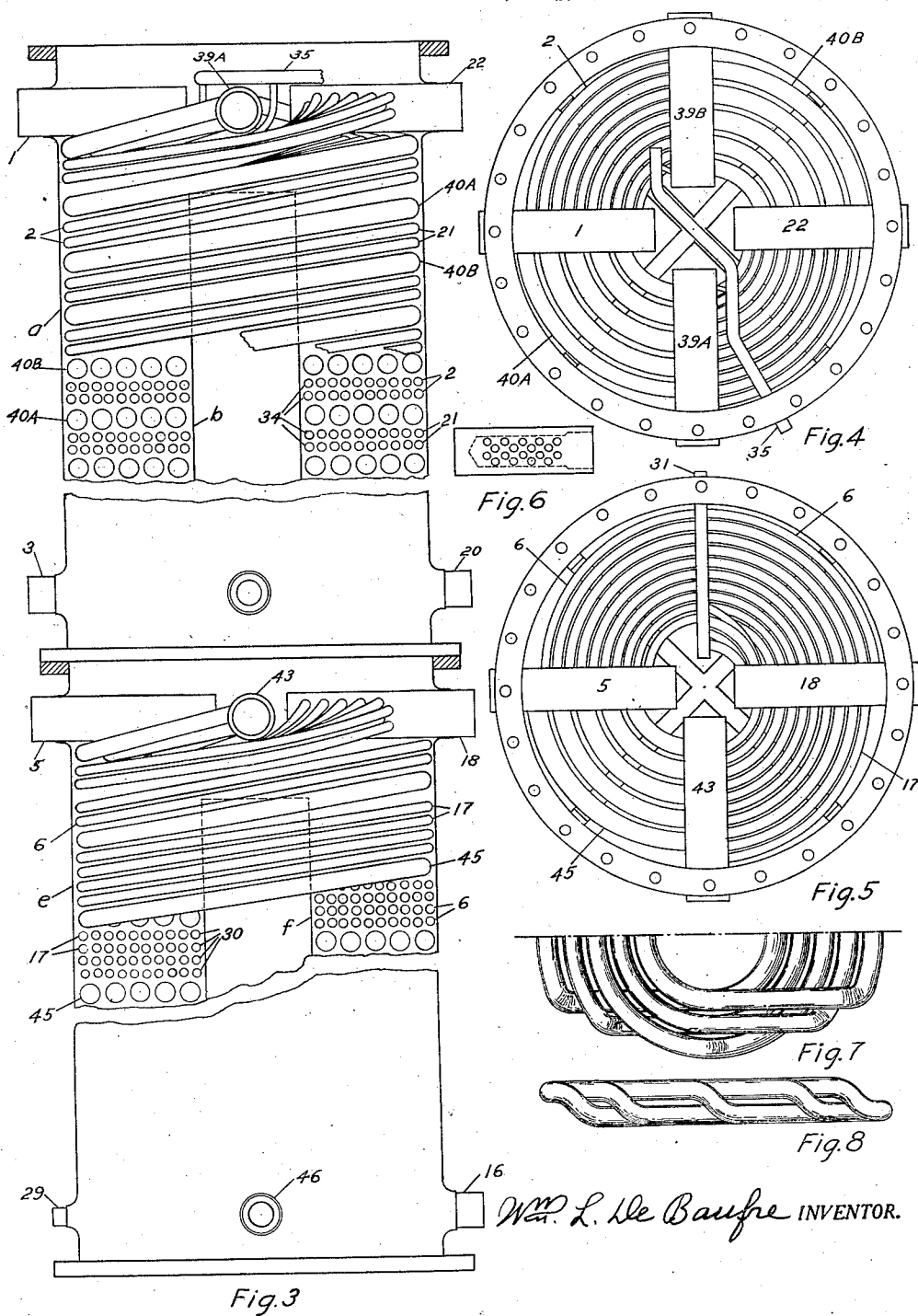
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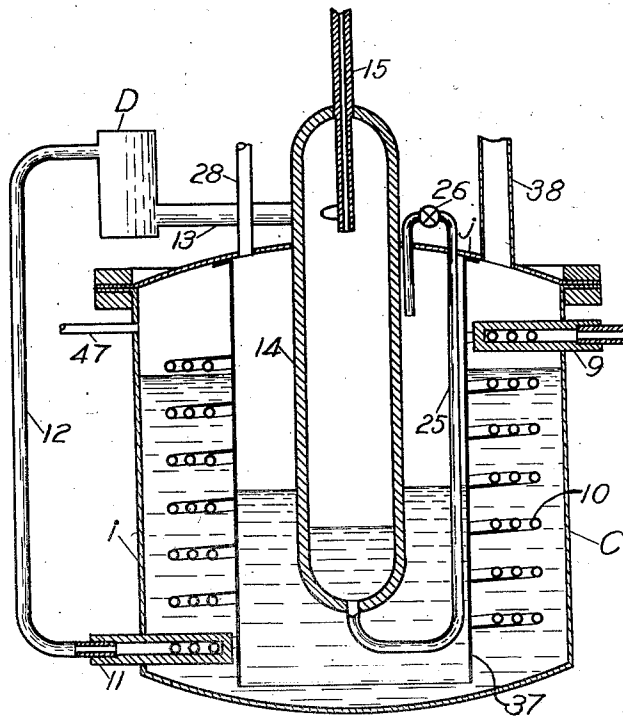


Fig. 9

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UNITED STATES PATENT OFFICE

1,961,202

APPARATUS FOR SEPARATING MIXED
GASES BY LIQUEFACTION

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Application March 17, 1932, Serial No. 599,475

11 Claims. (Cl. 62—122)

This invention relates to improvements in the art of separating mixed gases and is especially useful in the purification of helium containing nitrogen, oxygen and other gases having much higher boiling points than helium.

One of the primary objects of the invention is to secure helium of a higher purity than has heretofore been obtained by the liquefaction process.

A further object of the invention is to improve the efficiency of the apparatus and thereby reduce the power requirements to a minimum.

Another object of the invention is to increase the recovery of helium and reduce the possible loss of helium to a minimum.

Another object of the invention is to produce an apparatus which can readily be put into operation and which will operate for a considerable period without difficulty.

The above objects together with such other advantages as may hereinafter appear or are incident to the invention, are realized by the apparatus which I have illustrated in a preferred form in the accompanying drawings, wherein:

Fig. 1 is a front elevation of the apparatus;

Fig. 2 is a side elevation partly in section of the apparatus;

Fig. 3 is an elevation partly in section of the interchanger and liquefier;

Fig. 4 is a plan view of the interchanger;

Fig. 5 is a plan view of the liquefier;

Fig. 6 is a detail view of one of the manifolds used in the interchanger and liquefier;

Fig. 7 is a plan view in detail showing the method of reversing the positions of the helically coiled tubes in interchanger and liquefier;

Fig. 8 is an elevation of Fig. 7;

Fig. 9 is an elevation partly in cross-section of the purifier.

This patent application is supplementary to application No. 596,289 filed March 2, 1932, wherein the improved process was claimed as carried out in the herein described apparatus. Application No. 631,044, filed August 30, 1932, is a divisional application covering features shown but not claimed in this application.

Referring to Fig. 1 and Fig. 2, the apparatus comprises an interchanger A, a liquefier B, a purifier C, a mist collector D, and connecting pipes and valves, all suspended within a casing E filled with heat insulating material. A control board F serves for mounting the various pressure gages and liquid level gages convenient to the handles of the control valves. An expansion engine G may be provided, and the usual gas compressors, purifying towers and cylinders, etc.,

are required as in other processes for separating mixed gases by liquefaction.

Intercchanger A, as more fully shown in Fig. 3 and Fig. 4, comprises a cylindrical shell *a*, a concentric core *b*, and concentric helically coiled tubes 2, 21, etc., lying between shell *a* and core *b* and extending between radial manifolds 1, 3, etc., projecting through shell *a*. Tubes 40 are larger in diameter than tubes 2 and 21 for reasons stated later. A head *c*, shown in Fig. 2 but not in Fig. 3, is bolted to the upper end of interchanger A, while to the lower end is bolted liquefier B. Intercchanger A is suspended by bolts *d* from casing E.

Liquefier B, as shown in Fig. 3 and Fig. 5, comprises a cylindrical shell *e*, a concentric core *f*, and concentric helically coiled tubes 6, 17, etc., lying between shell *e* and core *f* and extending between radial manifolds 5, 16, etc., projecting through shell *e*. Tubes 45 are larger in diameter than tubes 6 and 17. A head *g* shown in Fig. 2 but not in Fig. 3, is bolted to the lower end of liquefier B.

Purifier C is suspended by bolts *h* from liquefier B, see Fig. 2. As shown more in detail in Fig. 9, purifier C consists of a cylindrical shell *i* closed at the lower end and having a head *j* on the upper end. A flask 14 is suspended in head, or cover, *j* and a partition 37 attached to cover *j* divides the space between flask 14 and shell *i* into an inner and an outer compartment. Helically coiled tubes 10 in the outer compartment extend between manifolds 9 and 11.

The casing E, Fig. 1 and Fig. 2, is made up of a structural steel framework of sufficient strength to support the weights of interchanger A, liquefier B, purifier C and mist collector D, all these being hung by bolts *d*. To the framework are fastened steel sheets, and the space between the steel sheets and the pieces of equipment suspended within is filled with a heat insulating material, such as rock wool.

The impure helium to be purified is first compressed to a pressure from 1500 to 3000 lb. per sq. in., dried and all traces of carbon dioxide removed in apparatus not shown in the drawings. The compressed impure helium is then introduced into manifold 1, Fig. 1, Fig. 3 and Fig. 4. From manifold 1, the compressed impure helium enters a number of small tubes 2 and flows therein down through interchanger A to manifold 3. Flowing thence through pipe 4, the impure helium, cooled to about 165 Kelvin, enters manifold 5 of liquefier B. From manifold 5, the impure helium enters a number of small tubes 6 and flows down through

liquefier B to manifold 7. The impure helium leaves manifold 7 through pipe 8 with a temperature of 100 Kelvin or less.

The impure helium then enters manifold 9 in purifier C, Fig. 9, flows down through a number of small tubes 10 to manifold 11 and leaves by pipe 12 cooled about to 75 Kelvin. At this very low temperature, most of the impurities are liquefied and a portion exists as mist in the helium gas. The helium gas containing this mist is passed through mist collector D where most of the mist is concentrated into liquid drops. The helium gas containing liquid drops of impurities and any remaining mist enters flask 14 through pipe 13. In flask 14, nearly complete separation occurs of all liquefied impurities from the helium gas.

The purified helium gas leaves flask 14 through pipe 15 and enters manifold 16 of liquefier B. It then passes up through a number of small tubes 17 to manifold 18 and leaves through pipe 19 at a temperature of about 145 Kelvin. The purified helium then enters manifold 20, flows up through tubes 21 in interchanger A and leaves through manifold 22 warmed nearly to atmospheric temperature. Valve 23 on the helium return pipe enables the pressure throughout the helium cycle just described to be controlled as desired.

The amount of liquefied impurities accumulated in flask 14 is shown by liquid level gage 24. These liquefied impurities, containing a small amount of absorbed helium gas, are discharged through pipe 25 and throttle valve 26, see Fig. 9. They are throttled from the pressure of 1500 to 3000 lb. per sq. in. within flask 14 to say 22 lb. per sq. in. in the annular space surrounding the lower portion of flask 14 in purifier C. At the low pressure after throttling, most of the absorbed helium gas is released. A small portion of the liquefied impurities is evaporated by the throttling action. The absorbed helium gas released and the liquid evaporated by throttling accumulate in the top of the annular space surrounding flask 14 and depress the liquid level in this annular space. The liquid level therein is shown by liquid level gage 27.

The mixture of helium gas and saturated vapor from the liquefied impurities is withdrawn from the annular space surrounding flask 14 through pipe 28 and returned to manifold 29 in liquefier B. Passing up through tubes 30, the mixture is warmed to about 145 Kelvin by the time it reaches manifold 31. It leaves through pipe 32 and enters manifold 33 in interchanger A. In tubes 34, the mixture is warmed nearly to room temperature by the time it reaches manifold 35 and leaves by a pipe containing valve 36 for regulating the flow of this recovered helium and hence the liquid level indicated on liquid level gage 27.

The liquefied impurities at a temperature of about 75 Kelvin from the inner compartment of purifier C immediately surrounding flask 14, are forced down under partition 37 and rise in the outer compartment of purifier C, flowing up over tubes 10. Heat transfer from the impure helium within tubes 10 first raises the temperature of the liquefied impurities to the temperature of vaporization corresponding to the pressure in the outer compartment and then vaporizes the liquid at this temperature. If the pressure in the outer compartment is say 22 lb. per sq. in., the temperature of vaporization will be over 80 Kelvin, the exact value depending upon the composition of the impurities. The liquid level in the outer compartment of purifier C is shown on liquid level gage 50.

The vaporized impurities return to liquefier B through pipe 38. In liquefier B and interchanger A, the vaporized impurities could be warmed nearly to room temperature in coils provided for the purpose. In the plant shown, however, the impurities are commingled with the air or other refrigerative medium used in a separate cycle to furnish the refrigeration to balance heat leak and other refrigeration requirements. The refrigeration cycle will now be described.

Air, nitrogen or other suitable working medium is compressed to say 500 lb. per sq. in., dried and all traces of carbon dioxide removed in apparatus not shown. The purified compressed air is then introduced into manifolds 39A and 39B. Passing down through tubes 40A and 40B in interchanger A, the compressed air is cooled to say 165 Kelvin by the time it reaches manifolds 41A and 41B. These manifolds are joined to an expansion engine G and to manifold 43 in liquefier B by means of piping 42. All or most of the compressed air passes to the expansion engine where it is expanded in doing work to a pressure somewhat above atmospheric pressure and then exhausted through pipe 44.

A small portion of the compressed air enters manifold 43 from piping 42 and is liquefied within tubes 45 joining manifolds 43 and 46 in liquefier B. Manifold 46 is connected to the upper part of the shell of purifier C by pipe 47 containing throttle valve 48. The liquid air accumulates within tubes 45 in liquefier B. At starting of the plant or whenever it is desired to augment the liquid within the outer compartment of purifier C, throttle valve 48 is opened and liquid air flows through pipe 47 from liquefier B to purifier C; that is, liquid air from the refrigeration cycle commingles with the liquefied impurities separated from the helium. The commingled liquids are vaporized by heat transfer from the impure helium within tubes 10 in purifier C. The resulting vapor leaves purifier C through pipe 38 and enters the shell of liquefier B.

The commingled impurities separated from the helium and the air from the refrigeration cycle pass up through liquefier B and interchanger A over all the tubes therein, and finally leave through pipe 49 containing an automatic pressure relief valve not shown. The control chamber of this automatic pressure relief valve is connected by a pipe to the top of the outer compartment of purifier C. By proper adjustment of the automatic pressure relief valve, any desired pressure can be maintained in the outer compartment of purifier C.

In interchanger A, helically coiled tubes 40A and 40B for the compressed air refrigerant are about twice the diameter of tubes 2 and 21 for the helium. This is done to reduce difficulties due to condensing and freezing of water vapor in the compressed air which is compressed to 500 to 600 lb. per sq. in. only as compared with 1500 to 3000 lb. per sq. in. for the helium. The higher the pressure to which air, helium or any other gas is compressed, the less is the quantity of water vapor remaining in a given mass of gas at a given temperature. This is equally true after the gas has been dried by passing over a chemical drying agent, which reduces the partial pressure of the water vapor to approximately the same value less than the saturation pressure corresponding to the temperature. Consequently, the compressed and dried air at 500 to 600 lb. per sq. in. carries considerably more water vapor into the interchanger than an equal weight

of compressed and dried helium at 1500 to 3000 lb. per sq. in. Although a lower rate of heat transfer occurs with tubes of larger diameter, it is desirable to use the larger diameter tubes for the gas of lower pressure in order to reduce any difficulties with moisture and ice. Another advantage of minor importance is the lower pressure drop with gas flow through larger diameter tubes, which becomes of greater importance the lower the gas pressure.

Liquefier B is of similar construction to that of interchanger A. Being of substantially the same diameter, these two parts of the apparatus can be bolted together to form one unit. Liquefier B is thus a continuation of interchanger A, and under certain conditions liquefier B can be built as one piece of equipment with interchanger A. With the manifolds for the helically coiled tubes projecting through the shells of interchanger A and liquefier B, these latter can be readily connected together, their heads attached thereto and piping connections readily made to these manifolds.

By reversing as shown in Fig. 7 and Fig. 8, the positions of the helically coiled tubes in interchanger A and liquefier B, so that the innermost tube becomes the outermost tube, the outermost tube becomes the innermost tube, and intermediate tubes are similarly interchanged, it becomes possible to make all tubes in any one radial row of substantially the same length between manifold. Consequently, the resistance to flow of fluid will be substantially the same through all tubes and the fluid flowing through any group of tubes will be equally divided among all tubes of the group. Also, the outside surface of each tube will be subjected to the flow of an equal quantity of flowing fluid. It therefore follows that heat transfer for all tubes will occur between the same quantities of fluids flowing within and without the tubes. The interchanger or liquefier as a whole will therefore be much more effective than if no reversals in tube position were made. In order to be most effective, an odd number of reversals, such as one, three, five, etc., should be employed, and these reversals should be made at equal intervals along the interchanger or liquefier. Without these reversals in tube positions, much more fluid would flow through the innermost tube than through the outermost tube. Also, the outside of the outermost tube would be subjected to much more fluid than the innermost tube. Consequently, the conditions for heat transfer would be very unequal with low efficiency for the unit as a whole.

The cores *b* and *f* are used in interchanger A and liquefier B respectively because it is not advisable to coil the tubes therein to a very small radius by reason of flattening the tubes if this is attempted. The cores fill the cylindrical spaces left within the coils of minimum permissible radius. If cores were not provided, most of the fluid returning within the shells would flow through these cylindrical spaces. The cores force the returning fluid to flow over the coils in intimate contact therewith.

The tubes in each radial row are closely spaced between the shell and the core in order to obtain a high rate of heat transfer through the tube wall. Where the tubes of one radial row enter a manifold, they are arranged in two staggered rows as shown in Fig. 6 in order to permit a manifold of reasonable thickness to be used. The tubes from two radial rows enter the mani-

fold in Fig. 6, which represents a manifold as used for the high pressure helium. The single rows of larger diameter tubes used for the compressed air are similarly arranged in two staggered rows where they enter the manifolds.

The suspension of purifier C from liquefier B enables interchanger A, liquefier B and purifier C all to be suspended within casing E. By making interchanger A, liquefier B and purifier C of substantially the same outside diameter, casing E can be of simple construction and ample heat insulation can be provided by filling the space between the casing and the interchanger, liquefier and purifier with some good insulating material, such as rock wool. Also, with this arrangement, the piping connections are comparatively simple and most of the pipes extending through the casing connect to the upper end of the interchanger where there is practically no relative motion between casing E and the points to which the pipes are attached within the casing.

The arrangement of purifier C as shown was adopted in order to reach the lowest possible temperature and to cool the impure helium as nearly as possible to that temperature. In order to reach the lowest possible temperature, it is necessary to protect from heat leak, the impurities throttled from the flask in which separation has occurred. Otherwise, such heat leak would vaporize some of the impurities and thereby increase the partial pressure of the vapor of these impurities above the remaining liquefied impurities. As the temperature of the remaining liquefied impurities corresponds to the partial vapor pressure of these impurities, any heat leak would thus raise the lowest possible temperature. Such heat leak is minimized in the apparatus shown by surrounding the compartment in which the absorbed helium is released and some of the liquid impurities are vaporized by throttling, with another compartment at nearly the same temperature. Constructionally, these inner and outer compartments are provided by means of partition 37 suspended from the cover of purifier C.

Coils 10 are placed in the outer compartment of purifier C and so connected that the impure helium flows down through these coils. As the helium approaches the lowermost coils, it comes into indirect contact with the liquid impurities cooled to the lowest possible temperature within the inner compartment. Consequently, the impure helium is cooled to a temperature closely approaching the lowest possible temperature. The purity reached by the helium is determined by the temperature to which it is thus cooled and to the effectiveness of separation of the liquefied impurities from the helium remaining as a gas.

The location of the lower end of flask 14 within the compartment into which the liquefied impurities are throttled, maintains the lower end of flask 14 at the lowest temperature in the system. There is therefore no tendency for the liquefied impurities within flask 14 to vaporize and thus reduce the purity of the helium gas from which they had previously been separated. Such vaporization not only adds these vapors to the purified helium but also tends to add thereto some mist of the liquefied impurities produced by boiling of the liquefied impurities.

While the vaporized impurities separated from the helium can be returned separately through coils in liquefier B and interchanger A, it is of advantage to commingle these impurities with

the refrigerant returning through the liquefier and the interchanger. Not only is the cost of these extra coils saved, but less material must be cooled down to operating temperatures and a greater mass of fluid flows up over the several coils in the liquefier and interchanger with resultant better heat transfer. During operation, the plant may run with all the refrigerant passing through the expansion engine and with none throttled through valve 48 into purifier C. If the liquid level in the outer compartment drops, however, then valve 48 may be partly opened to admit liquid air into this compartment. It will be noticed that this liquid air is admitted into the top of the outer compartment of purifier C in order that the impure helium within coils 10 comes first into indirect contact with liquid air or with a mixture of liquid air and impurities before it finally comes into indirect contact with the liquefied impurities from the inner compartment of purifier C at a much lower temperature than that of the liquid air.

The continued addition to the refrigeration cycle of the impurities removed from the impure helium processed, is more than ample to make up for any losses by leakage from the refrigeration cycle. As these impurities are free of moisture and carbon dioxide, the consumption of chemicals for drying and removal of carbon dioxide in the refrigeration cycle is appreciably reduced over that required when the make up is atmospheric air.

Should, for any reason, the liquid level in the inner compartment of purifier C be permitted to become so low as to discharge vapor containing helium into the outer compartment, this helium is not necessarily lost but may be recovered by pumping the gas from the refrigeration cycle into the impure helium storage and then processing it as impure helium.

By compressing the air used as a refrigerant to a higher pressure than 500 to 600 lb. per sq. in., say 1000 to 3000 lb. per sq. in., the Joule-Thomson effect will be sufficient to obtain the necessary refrigeration without expansion engine G. The modification of the interchanger and liquefier necessary with this variation in the refrigeration cycle, is contemplated in the wording of certain of the claims appended hereto.

Separate interchangers might be used for the air of the refrigeration cycle and for the impure helium, the purified helium and the impurities separated therefrom. Since both the impure helium and the purified helium are under substantially the same high pressure, the shell of the second interchanger would have to be designed for this high pressure for containing either the impure helium cooled or the purified helium warmed. By the combination adopted, both the impure helium cooled and the purified helium warmed flow through tubes and manifolds which can easily be designed to withstand very high pressures. The cooling and warming is effected by heat interchange relation with the refrigerant being warmed. This refrigerant is under a pressure but little above atmospheric pressure, so that the shell of the combination interchanger is subjected to a low pressure only.

The value of raising the purity of helium for airships from 98 to 99.5 per cent is indicated by the fact that this increase in purity raises the cargo carrying capacity of the U. S. S. *Akron* by about three tons. When the purity of the helium in airships has dropped to 90 percent by reason of air infiltration through the fabric, it has been

customary to repurify the helium. The period between times of repurification is increased nearly 20 per cent by the use of 99.5 per cent helium as compared with 98 per cent helium. By means of the apparatus herein described and claimed, these advantages are obtained along with decreased power consumption due to the possibility of operating with 2000 lb. per sq. in. helium pressure as compared with 2500 lb. per sq. in. previously used, and to the need of a refrigeration cycle of about one-half the capacity of that previously required. The consumption of chemicals for drying and removing carbon dioxide from the refrigeration cycle is reduced by reason of the make-up for leakage from this cycle being entirely free of moisture and carbon dioxide. In addition, a safeguard is provided against loss of helium during operation.

I claim:

1. In an apparatus for separating impurities by selective liquefaction from a gas having a much lower boiling point than that of any of the said impurities and utilizing a refrigerant, a casing, an interchanger suspended within said casing for cooling said gas and said refrigerant, a liquefier suspended from said interchanger for further cooling said gas and liquefying a portion of said refrigerant, a purifier suspended from said liquefier for purifying said gas, and piping connections between said interchanger and said liquefier and between said liquefier and said purifier for conveying gas and refrigerant.

2. An apparatus for separating impurities by selective liquefaction from a gas having a much lower boiling point than that of any of the said impurities, comprising means for supplying thereto the said gas, means for supplying thereto a refrigerant for balancing heat leak and other refrigeration requirements, an interchanger for cooling said gas and said refrigerant, a liquefier for further cooling said gas and liquefying a portion of said refrigerant, a purifier for purifying said gas, and pipe connections between said interchanger and said liquefier and between said liquefier and said purifier for conveying gas and refrigerant.

3. An apparatus for separating impurities by selective liquefaction from a gas having a much lower boiling point than that of any of the said impurities, comprising means for supplying thereto the said gas, means for supplying thereto a refrigerant for balancing heat leak and other refrigeration requirements, an interchanger for cooling said gas and said refrigerant by returning commingled impurities and refrigerant, a liquefier for further cooling said gas and liquefying a portion of said refrigerant by returning commingled impurities and refrigerant, a purifier for further cooling said gas by liquid impurities and liquid refrigerant and for separating impurities from said gas, and pipe connections between said interchanger and said liquefier and between said liquefier and said purifier for conveying gas and refrigerant.

4. An apparatus for purifying compressed gas, comprising means for supplying thereto the said compressed gas, means for supplying thereto a compressed gaseous refrigerant for balancing heat leak and other refrigeration requirements, an interchanger for cooling said compressed gas and said compressed gaseous refrigerant by returning refrigerant at a lower pressure, a liquefier for further cooling said compressed gas and liquefying a portion of said compressed gaseous refrigerant by returning refrigerant at a lower

pressure, a purifier for further cooling said compressed gas by liquefied impurities separated therefrom and for separating said impurities, means for reducing the pressure of said compressed gaseous refrigerant, and piping between said means and said liquefier, between said liquefier and said purifier and between said interchanger and said liquefier for conveying gas and impurities and refrigerant.

5. In an apparatus for separating mixed gases by liquefaction, an interchanger comprising a shell, a core, radial manifolds projecting through said shell, and helically coiled tubes lying in radial rows between said shell and said core and extending between said radial manifolds, the coiled tubes in one radial row being staggered where they enter the radial manifold.

6. In an apparatus for separating mixed gases by liquefaction wherein high and low pressure fluids are in heat interchange with another fluid, an interchanger comprising a shell, a core, high pressure manifolds projecting through said shell for conveying the high pressure fluid, low pressure manifolds projecting through said shell for conveying the low pressure fluid, coiled tubes of small diameter lying between the shell and the core and extending between the high pressure manifolds, and coiled tubes of larger diameter lying between the shell and the core and extending between the low pressure manifolds.

7. In an apparatus for separating mixed gases by liquefaction, an interchanger comprising a shell, a core, and helically coiled tubes lying between said shell and said core and extending between manifolds, the said helically coiled tubes being reversed in radial position at intervals in the length of the interchanger so as to make all tubes of substantially the same length.

8. In an apparatus for separating impurities by selective liquefaction from a gas having a much lower boiling point than that of any of the said impurities, a vessel in which the liquefied impurities are separated from the gas, a compartment wherein the liquefied impurities throttled from the said vessel are subjected to the pressure of the absorbed gas released and the

liquid impurities evaporated by throttling, and a second compartment wherein the remaining liquid impurities are warmed and evaporated by heat transfer from the said gas in cooling the gas nearly to the temperature of the liquefied impurities after throttling.

9. In an apparatus for separating impurities by selective liquefaction from a gas having a much lower boiling point than that of any of the said impurities, a vessel in which the liquefied impurities are separated from the gas, a compartment surrounding said vessel wherein the liquefied impurities throttled from the said vessel are subjected to the pressure of the absorbed gas released and the liquid impurities evaporated by throttling, and a second compartment wherein the remaining liquid impurities are warmed and evaporated by heat transfer from a fluid to be cooled.

10. In an apparatus for separating impurities by selective liquefaction from a gas having a much lower boiling point than that of any of the said impurities, a vessel in which the liquefied impurities are separated from the gas, a compartment wherein the liquefied impurities throttled from the said vessel are subjected to the pressure of the absorbed gas released and the liquid impurities evaporated by throttling, a second compartment surrounding the before mentioned compartment wherein the remaining liquid impurities are warmed and evaporated by heat transfer from a fluid to be cooled.

11. In an apparatus for separating impurities by selective liquefaction from a gas having a much lower boiling point than that of any of the said impurities, a purifier comprising a shell and a cover, a flask suspended in said cover wherein the liquefied impurities are separated from the gas, a partition attached to said cover and dividing the space between the said flask and the said shell into two compartments, tubes in the outer compartment for conveying the gas containing the impurities and means for throttling the liquefied impurities from the said flask into the inner compartment.

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