

March 1, 1966

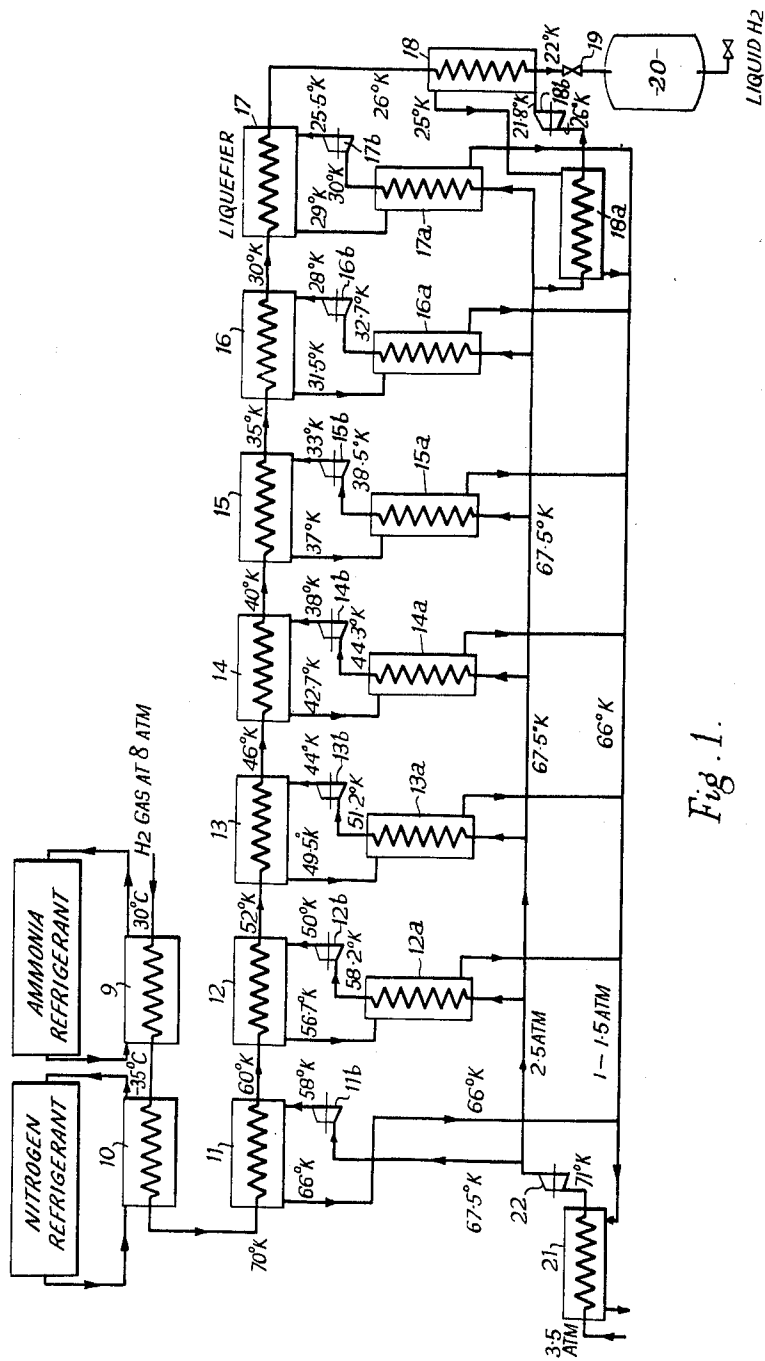
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LIQUEFACTION OF GASES

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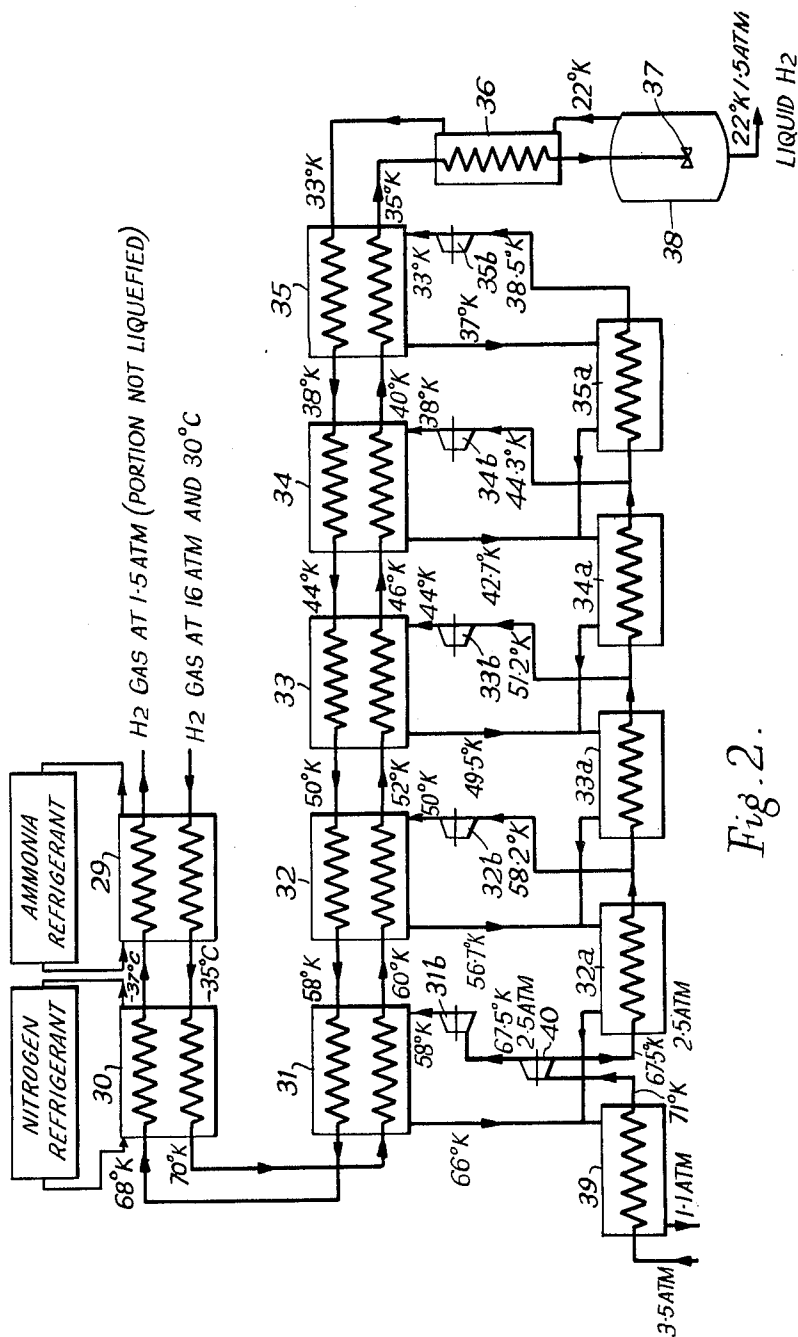


Fig. 2.

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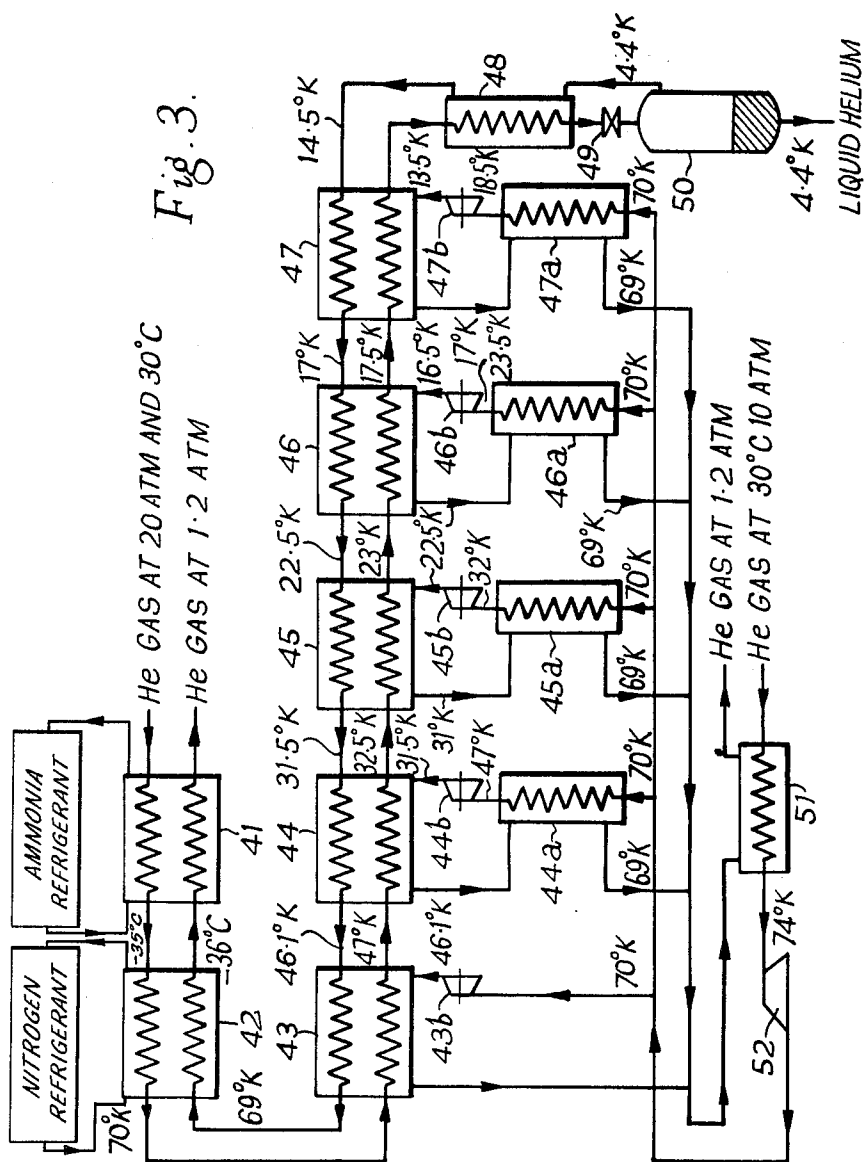
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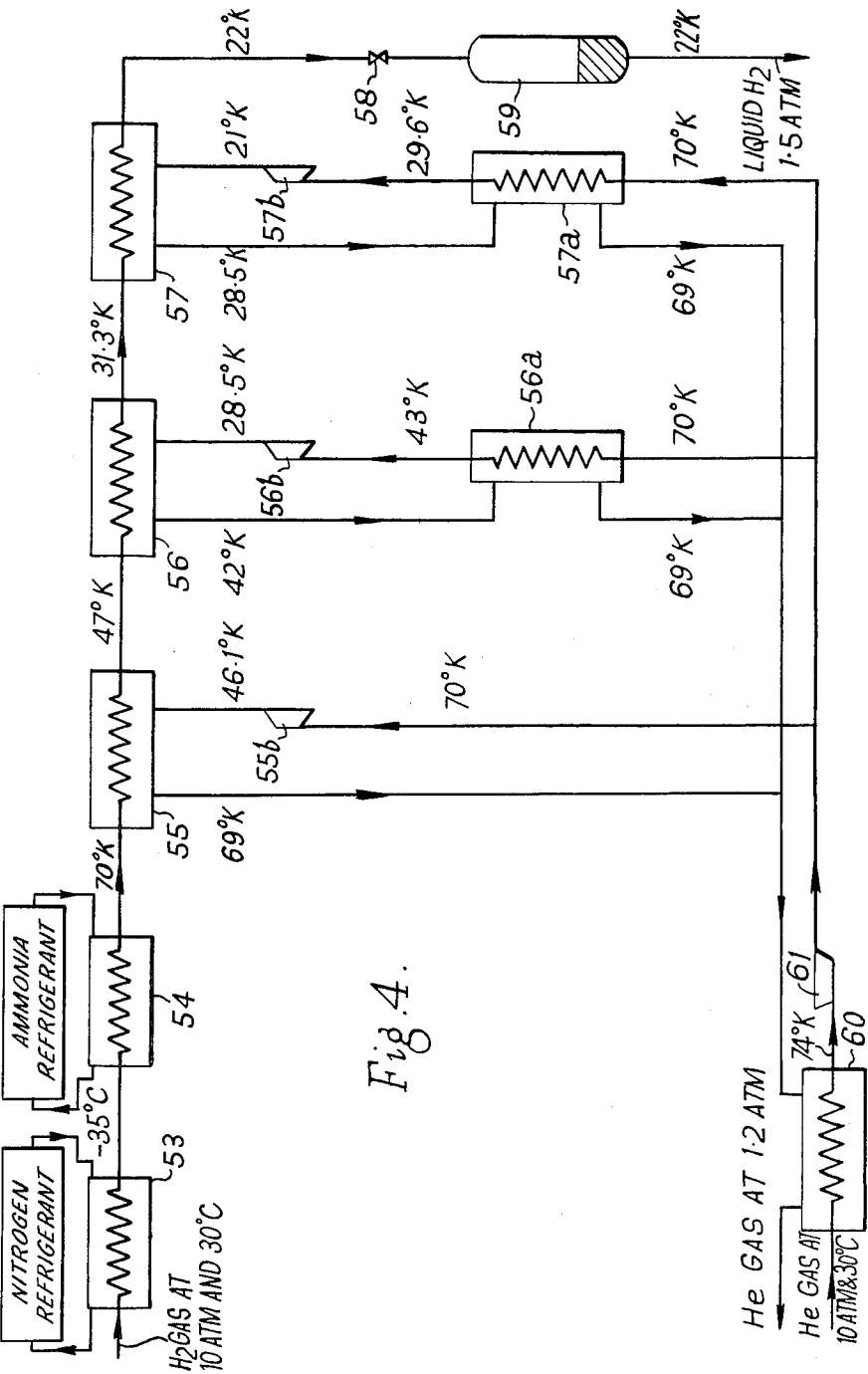
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LIQUEFACTION OF GASES

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9 Claims. (Cl. 62—9)

This invention relates to methods of cooling gases down to very low temperatures i.e. to temperatures appreciably below 70° K. It is primarily concerned with the liquefaction of gases such as hydrogen and helium which have a liquefaction temperature well below 70° K.

In the known method of liquefying hydrogen, hydrogen at 170 atmospheres is cooled first with liquid nitrogen boiling at atmospheric pressure and then with liquid nitrogen under vacuum down to a temperature of 65° K. It is then expanded to 1.5 atmospheres with the result that about 25% of the hydrogen is liquefied, making use of the Joule-Thomson effect. Because of the high pressures involved and for other reasons, this method is not suitable for use on a commercial scale.

The invention provides a method whereby a gas at a moderately elevated pressure, i.e. a pressure not substantially in excess of 30 atmospheres, may be cooled down to its liquefaction temperature or to a temperature at which partial liquefaction may be effected by expansion, making use of the Joule-Thomson effect.

According to the invention a method of cooling a gas to a temperature well below 70° K. comprises passing the gas at a moderately elevated pressure and at a temperature of the order of 65 to 70° K. through a series of cooling zones and cooling the gas in each zone with a separate stream of a coolant gas, which has a boiling point not higher than that of the gas being cooled and has finally been cooled to the temperature required by expansion through a turbine. The temperature of the coolant gas leaving each zone is lower than that of the inlet gas to the turbine associated with that zone. Normally the coolant gas expanded through the turbines will be expanded from a pressure lower than that of the gas being cooled. The coolant gas for each of the second and following cooling zones, is preferably cooled by the coolant gas leaving that cooling zone prior to its expansions through the turbine.

Cooling of the gas down to 65° to 70° K. may be effected in known fashion, for example by ammonia refrigeration followed by cooling with nitrogen.

The method of the invention may be applied to the liquefaction of hydrogen, using hydrogen or helium as the coolant gas or to liquefying helium using helium as the coolant gas.

In applying the method to the liquefaction of hydrogen, the hydrogen feed may suitably be at 65° to 70° K. and a pressure of from 2 to 20 atms. The hydrogen may be liquefied by cooling down to its liquefaction temperature, providing its pressure is below the critical pressure, or to a temperature at which the Joule-Thomson effect may advantageously be used and liquefaction of part effected by expansion.

In applying the method to the liquefaction of helium, the helium feed may suitably be at a temperature of 65° to 70° K. and at a pressure of from 5 to 30 atms. Because of the very low critical temperature and pressure of helium, the final stage of cooling and liquefaction is by expansion making use of the Joule-Thomson effect.

In the method of liquefying hydrogen, in which cooling down to the liquefaction temperature or below is effected by refrigeration, the hydrogen at a pressure not exceeding 12.5 atmospheres, and suitably at 8 atmospheres, and at a temperature of 70° K. may be cooled in 7 stages in heat exchangers down to 26° K. at which temperature it is liquid, its liquefaction temperature being

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30° K. and then subcooled to 22° K. in an eighth stage and the liquid at 22° K. may then be expanded to 1.5 atmospheres. Each stage reduces the temperature by an amount varying from 10° K. to 4° K. and starting from 70° K. the successive stages of cooling are down to 60° K., 52° K., 46° K., 40° K., 35° K., 30° K., 26° K. and 22° K. respectively. In each stage cooling may be effected by hydrogen as the coolant gas cooled by expansion from 2.5 atmospheres to 1 to 1.5 atmospheres through a turbine to a temperature about 2° K. below that to which cooling is to be effected in the relevant stage, the cooling gas leaving the heat exchanger of each stage at a temperature below that at which the gas enters the turbine associated with that stage. In each cooling stage except the first, the cooling gas entering the turbine is passed through a precooler, which is cooled by the coolant gas leaving the heat exchanger of that particular stage. The main cooling gas feed is supplied at about 71° K. and at a pressure of 3.5 atmospheres. It is then expanded to 2.5 atmospheres through a turbine and thus cooled to 67.5° K. The cooling stream at 67.5° K. is fed in parallel to all the cooling stages, thus it is fed to the turbine of the first stage in which it is expanded to 1 to 1.5 atmospheres and to the precoolers of each of the succeeding stages. The return gas which leaves the first heat exchanger and each of the precoolers at a temperature of 66° K. is collected and passed through a heat exchanger to the compressor in which it is recompressed to 3.5 atmospheres. It is then returned, via the last mentioned heat exchanger in which it is cooled to about 71° K., to the cooling system.

In the alternative method in which the hydrogen gas is cooled down to 35° K. and then partially liquefied by expansion, the first five stages of cooling from 70° K. down to 35° K. are similar to those described above, though the hydrogen to be cooled may advantageously be at a higher pressure, suitably 16 atmospheres. The hydrogen at 35° K. may then be passed through a further heat exchanger and then to the expansion vessel in which it is expanded to 1.5 atmospheres and partially liquefied. The portion not liquefied is passed back as a coolant through the last mentioned heat exchanger and then in series through the heat exchangers of cooling stages 5 to 1 and also through the nitrogen and ammonia refrigeration systems employed in cooling down to 70° K. About 41.5% of the hydrogen fed into the system is liquefied on expansion under the conditions specified above. An appreciable proportion of the power used in either method is recovered from the turbines employed in the cooling stages.

In the liquefaction of hydrogen, helium may be used as the coolant gas, the cycle being appropriately modified. Thus hydrogen at a pressure of 10 atmospheres and a temperature of 70° K. may be cooled with helium in three stages to 22° K., the successive stages being down to 47° K., 31.3° K. and 22° K. The helium coolant gas is at 10 atmospheres and is cooled from room temperature to 74° K. by the return stream of coolant gas. It then passes through a turbine in which it is expanded from 10 to 7 atmospheres and is cooled to 70° K. The coolant gas is fed to each of the three cooling stages in each of which it is finally cooled by expansion through a turbine from 7 to 1.2 atmospheres to a temperature about 1° K. below that to which the hydrogen stream is to be cooled in the relevant stage. In the second and third stages the helium is precooled before entering the turbine by passage through a heat exchanger cooled by the coolant helium returning from the respective cooling stage.

In applying the method of the invention to the liquefaction of helium using helium as coolant, helium at a moderately elevated pressure not substantially exceeding

30 atmospheres and at a temperature of the order of 65° to 70° K. may be cooled by the method of the invention until it reaches a temperature, for example 15° K., from which it may be cooled by return gases in a final heat exchanger to a temperature at which the Joule-Thomson effect may advantageously be used and liquefaction of part of the helium be effected by expansion, the remainder of the helium being returned through the final heat exchangers and the preceding heat exchangers.

In this method, the helium stream to be liquefied, which may advantageously be at 20 atmospheres pressure and has been cooled in known fashion to 70° K. by refrigeration with ammonia and nitrogen, may be cooled in five stages in heat exchangers down to 15° K. It then passes through a further heat exchanger in which it is cooling by returning gas to 7° K. and then through a Joule-Thomson valve where the pressure falls to about 1.5 atmospheres and 11% of the stream is liquefied. The remainder of the stream is returned through all the heat exchangers and the ammonia and nitrogen refrigeration stages back to the compressor. The coolant stream of helium, at 10 atmospheres, is cooled from room temperature to 74° K. in a heat exchanger by the return stream of coolant gas from all five of the cooling stages. It then passes through a turbine to be expanded from 10 to 7 atmospheres, and thus cooled to 70° K. The coolant gas is fed to each of the five coolant stages. Each stage reduces the temperature of the helium stream being cooled by an amount varying from 23° K. to 2.5° K. and, starting from 70° K., the successive stages of cooling are down to 47° K., 32.5° K., 17.5° K. and 15° K. In each stage cooling is obtained by expanding the helium gas from 7 atmospheres to 1.2 atmospheres through a turbine to a temperature about 1° K. below that to which cooling of the helium stream is to be effected in that stage. In each cooling stage except the first, the cooling gas entering the turbine is passed through a heat exchanger cooled by the gas leaving the heat exchanger of that stage.

Methods for liquefying hydrogen and helium in accordance with the invention are illustrated, by way, of example, diagrammatically in the accompanying drawings, in which:

FIGURE 1 illustrates a method of cooling hydrogen, using hydrogen as coolant, down to the liquefaction temperature.

FIGURE 2 illustrates a method of cooling hydrogen, using hydrogen as coolant, down to 35° K. by refrigeration and then further cooling and liquefying by expansion using the Joule-Thomson effect,

FIGURE 3 illustrates a method of cooling helium, using helium as coolant, down to 15° K. by refrigeration and then, after further cooling to 7° K., liquefying by expansion using the Joule-Thomson effect,

FIGURE 4 illustrates a method of cooling hydrogen, using helium as coolant, down to the liquefaction temperature.

Referring to FIGURE 1, 9 and 10 represent the preliminary refrigeration stages in which ammonia and nitrogen respectively are used for cooling hydrogen at 8 atmospheres down to 70° K. 11, 12, 13, 14, 15, 16 and 17 are the heat exchangers in which further cooling is effected in stages as shown down to 60° K., 52° K., 46° K., 40° K., 35° K., 30° K. and finally down to 26° K. which is below the liquefaction temperature. 18 is the heat exchanger for sub-cooling to 22° K. after which the liquid is expanded through valve 19 to 1.5 atmospheres and collected in container 20.

In the cooling circuit, hydrogen at 3.5 atmospheres and about 300° K. coming from the compressor is cooled by the return gas in heat exchangers 21 and then expanded through turbine 22 to 2.5 atmospheres and thus cooled to 67.5° K. The coolant at a pressure of 2.5 atmospheres and at a temperature of 67.5° K. is then passed in parallel through

(1) Turbine 11b leaving at 1 to 1.5 atmospheres and

at a temperature of 58° K. to pass through heat exchanger 11 and then back to heat exchanger 21 at a temperature of 66° K.

(2) Precooler 12a and turbine 12b entering the turbine 12b at a temperature of 58.2° K. and leaving at a temperature of 50° K. and a pressure of 1 to 1.5 atmospheres to pass through heat exchanger 12 from which it returns to precooler 12a at a temperature of 56.7° K. The coolant after passing through precooler 12a joins the return stream passing to heat exchanger 21 at 66° K.

(3) Precooler 13a and turbine 13b entering turbine 13b at a temperature of 51.2° K. and leaving at a temperature of 44° K. and a pressure of 1 to 1.5 atmospheres, the coolant then passing through heat exchanger 13 and returning to precooler 13a at a temperature of 49.5° K. and passing therefrom to the return stream passing to heat exchanger 21 at 66° K.

(4) Precooler 14a and turbine 14b entering turbine 14b at a temperature of 44.3° K. and leaving at a temperature of 38° K. and a pressure of 1 to 1.5 atmospheres, the coolant then passing through heat exchanger 14 leaving at a temperature of 42.7° K. and returning via precooler 14a to the return stream passing to heat exchanger 21 at 66° K.

(5) Precooler 15a and turbine 15b entering turbine 15b at a temperature of 38.5° K. and leaving at a temperature of 33° K. and a pressure of 1 to 1.5 atmospheres, the coolant then passing through heat exchanger 15 leaving at a temperature of 37° K. and returning via precooler 15a to the return stream passing to heat exchanger 21 at 66° K.

(6) Precooler 16a and turbine 16b entering turbine 16b at a temperature of 32.7° K. and leaving at a temperature of 28° K. and a pressure of 1 to 1.5 atmospheres, the coolant then passing through heat exchanger 16 leaving at a temperature of 31.5° K. and returning via precooler 16a to the return stream passing to heat exchanger 21 at 66° K.

(7) Precooler 17a and turbine 17b entering turbine 17b at a temperature of 30° K. and leaving at a temperature of 25.5° K. and a pressure of 1 to 1.5 atmospheres, the coolant then passing through heat exchanger 17 leaving at a temperature of 29° K. and returning via precooler 17a to the return stream passing to heat exchanger 21 at 66° K.

(8) Precooler 18a and turbine 18b entering turbine 18b at a temperature of 26° K. and leaving at a temperature of 21.8° K. and a pressure of 1 to 1.5 atmospheres, the coolant then passing through heat exchanger 18 leaving at a temperature of 25° K. and returning via precooler 18a to the return stream passing to heat exchanger 21 at 66° K.

Referring to FIGURE 2, 29 and 30 represent the preliminary ammonia and nitrogen refrigeration stages for cooling hydrogen at 16 atmospheres down to 70° K. Heat exchangers 31, 32, 33, 34 and 35 and their associated cooling circuits comprising turbine 31b, turbine 32b, turbine 33b, turbine 34b, and turbine 35b, respectively, operate in a similar fashion to heat exchangers 11 to 15 of the system illustrated in FIGURE 1 except that the heat exchangers 31 to 35 are additionally cooled by a return stream of hydrogen which is not liquefied in the subsequent expansion step. Furthermore the precoolers 32a, 33a, 34a and 35a are arranged in series in the present case, the feeds to the associated turbines 32b to 35b being taken off in parallel. The hydrogen at 16 atmospheres leaving heat exchanger 35 at 35° K. passes through heat exchanger 36 and then is expanded through valve 37 in chamber 38 to realise the Joule-Thomson effect. By this means it is cooled down to 22° K. and about 41.5% is liquefied. The non-liquefied gas is returned through heat exchanger 36 and then backwards in series through heat exchangers 35 to 31 and through the cooling stages 30 and 29.

The coolant gas which has passed through heat ex-

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changers 39 and turbine 40 at 2.5 atmospheres is then expanded through each of the turbines 31*b*, 32*b*, 33*b*, 34*b* and 35*b* to 1.1 atmospheres.

The temperature at different points of the system are shown in FIGURE 2.

Referring to FIGURE 3, 41 and 42 represent the preliminary ammonia and nitrogen refrigeration stages for cooling helium at 20 atmospheres down to 70° K. Heat exchangers 43, 44, 45, 46 and 47 and their associated cooling circuits comprising turbine 43*b*, turbine 44*b*, turbine 45*b*, turbine 46*b* and turbine 47*b* respectively, operate in a similar fashion to heat exchangers 31 to 35 of the system illustrated in FIGURE 2 and the heat exchangers 43 to 47 being additionally cooled by a return stream of helium which is not liquefied in the subsequent expansion step. The precoolers 44*a*, 45*a*, 46*a* and 47*a* are arranged in parallel, and are connected to the associated turbines as in the system illustrated in FIGURE 1. The helium is cooled in heat exchangers 43, 44, 45 and 46 respectively to 47° K., 32.5° K., 23° K. and 17.5° K. and leaves heat exchanger 47 at 15° K. It then passes through heat exchanger 48 in which it is further cooled by returning helium gas of the main gas stream to 7° K. and is then expanded through valve 49 into chamber 50 to realise the Joule-Thomson effect. The helium pressure falls to 1.5 atmospheres and its temperature to 4.4° K. and 11% of the helium stream is liquefied. The non-liquefied helium gas is returned through heat exchanger 48 to effect cooling therein and then backwards successively through heat exchangers 47 to 43 and cooling stages 42 and 41.

The coolant stream of helium is at 10 atmospheres and is cooled from room temperature to 74° K. in heat exchanger 51 by the return stream of coolant gas from all the cooling stages. It is then expanded to 7 atmospheres through turbine 52. The coolant helium stream at a pressure of 7 atmospheres and temperature 70° K. is passed through the five parallel stages described below.

(1) Turbine 43*b* leaving at 1.2 atmospheres and a temperature of 46.1° K. to pass through heat exchanger 43 and then return to heat exchanger 52 at a temperature of 69° K.

(2) Precooler 44*a* and entering turbine 44*b* at 47° K. and leaving at a temperature of 31.5° K. and pressure 1.2 atmospheres to pass through heat exchanger 44 from which it returns to precooler 44*a* at a temperature of 46.1° K. The coolant returns from precooler 44*a* to exchanger 52 at a temperature of 69° K.

(3) Precooler 45*a* and entering turbine 45*b* at 32° K. and leaving at a temperature of 22.5° K. and pressure 1.2 atmospheres to pass through heat exchanger 45 from which it returns to precooler 45*a* at a temperature of 31° K. The coolant returns from precooler 45*a* to exchanger 52 at a temperature of 69° K.

(4) Precooler 46*a* and entering turbine 46*b* at 23.5° K. and leaving at a temperature of 16.5° K. and pressure 1.2 atmospheres to pass through heat exchanger 46 from which it returns to precooler 46*a* at a temperature of 22.5° K. The coolant returns from precooler 46*a* to exchanger 52 at a temperature of 69° K.

(5) Precooler 47*a* and entering turbine 47*b* at 18.5° K. and leaving at a temperature of 13.5° K. and pressure 1.2 atmospheres to pass through heat exchanger 47 from which it returns to precooler 47 at a temperature of 17° K. The coolant returns from precooler 46*a* to exchanger 52 at a temperature of 69° K.

Referring to FIGURE 4, 53 and 54 represent the preliminary stages in which ammonia and nitrogen are used for cooling the liquefaction hydrogen stream at 10 atmospheres to 70° K. 55, 56 and 57 are heat exchangers in which further cooling is effected in stages down to 47° K., 31.3° K. and finally down to 22° K. at which stage the hydrogen is liquefied. 58 is the expansion valve where the pressure falls from 10 atmospheres to 1.5 atmospheres and the liquid gas is collected in vessel 59.

In the cooling circuit helium at 10 atmospheres and

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303° K. from the compressor is cooled by the return coolant gas in heat exchanger 60 and then expanded to 7 atmospheres through turbine 61. The coolant helium stream at a pressure of 7 atmospheres and temperature 70° K. is passed through the three parallel stages described below.

(1) Turbine 55*b* leaving at 1.2 atmospheres and a temperature of 46.1° K. to pass through heat exchanger 55 and then return to heat exchanger 60 at a temperature of 69° K.

(2) Precooler 56*a* and entering turbine 56*b* at 43° K. and leaving at a temperature of 28.5° K. and pressure 1.2 atmospheres to pass through heat exchanger 56 from which it returns to precooler 56*a* at a temperature of 42° K. The coolant returns from precooler 56*a* to exchanger 60 at a temperature of 69° K.

(3) Precooler 57*a* and entering turbine 57*b* at 29.6° K. and leaving at a temperature of 21° K. and pressure 1.2 atmospheres to pass through heat exchanger 57 from which it returns to precooler 57*a* at a temperature of 28.5° K. The coolant returns from precooler 57*a* to exchanger 60 at a temperature of 69° K.

The expansion turbines used in the cooling systems described above are preferably of the axial flow type and the heat exchangers are preferably of the plate-fin type.

All pressures referred to hereinbefore and in the claims are absolute pressures.

I claim:

1. A method of cooling a gas to a temperature substantially below 70° K. which comprises:

(a) passing the gas to be cooled, at an elevated pressure and at a temperature between about 65 and 70° K., serially through a plurality of separate, first cooling zones;

(b) separating a feed stream of coolant gas, at an elevated pressure, into a plurality of separate streams of coolant gas equal in number to the number of said first cooling zones;

(c) thereafter, passing each of said separate streams of coolant gas through a plurality of expansion turbines, each of which is operatively associated with one of said plurality of first cooling zones, to expand each of said separate streams of coolant gas and form an equal number of separate streams of expanded coolant gas;

(d) passing each of said separate streams of expanded coolant gas through one of said first cooling zones, in indirect heat exchange with said gas to be cooled as said gas to be cooled passes through said first cooling zones, to cool said gas to be cooled in each of said first cooling zones to a temperature lower than the temperature of said separate streams of coolant gas passing to said turbines and produce an equal number of separate streams of partially heated, expanded coolant gas;

(e) passing each of the second and subsequent of said partially heated, expanded streams of coolant gas through a plurality of second cooling zones, separate from one another and from said first cooling zones, to pre-cool the associated one of said separate streams of coolant gas passing to said turbines and produce an equal number of streams of additionally heated, expanded coolant gas;

(f) recombining said partially heated, expanded coolant gas from the first of said cooling zones and said separate streams of additionally heated, expanded coolant gas from said second cooling zones, to form a return stream of coolant gas;

(g) cooling the said feed stream of coolant gas with said return stream of coolant gas prior to the separation of said feed stream of coolant gas into said plurality of separate streams of coolant gas; and

(h) thereafter, passing said return stream of coolant gas to a compressor and thence to said feed stream of coolant gas.

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2. A method as claimed in claim 1, wherein the feed stream of coolant gas is expanded through an expansion turbine to a pressure lower than that of the gas to be cooled and prior to the separation of said feed stream of coolant gas into a plurality of streams of coolant gas.

3. A method as claimed in claim 1, wherein the gas to be cooled is cooled to 65° to 70° K. by refrigeration successively with ammonia and with nitrogen.

4. A method as claimed in claim 1, wherein the gas to be cooled is hydrogen and said hydrogen at a pressure of from 2 to 12.5 atmospheres is cooled down to at least its liquefaction temperature to liquefy the hydrogen, the said coolant gas being selected from the group consisting of hydrogen and helium.

5. A method as claimed in claim 4, wherein the said coolant gas is hydrogen and each separate stream of coolant gas is expanded through its corresponding turbine from a pressure lower than that of the hydrogen being cooled in the corresponding cooling zone.

6. A method as claimed in claim 1, wherein the gas to be cooled is hydrogen and said hydrogen at a pressure of from 2 to 20 atmospheres is cooled down to a temperature at which, on expansion, the Joule-Thomson effect may be used to liquefy part of the hydrogen, the said coolant gas being selected from hydrogen and helium.

7. A method as claimed in claim 6, wherein the cooled hydrogen is expanded and thus further cooled to liquefy part of the hydrogen and an additional final cooling stage is included prior to the expansion and liquefaction step, the coolant in this final stage being unliquefied hydrogen returned from the liquefaction step.

8. A method as claimed in claim 1, wherein the gas to be cooled is helium and said helium at a pressure of the order of 5 to 30 atmospheres is cooled down to a temper-

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ature at which, on expansion, the Joule-Thomson effect may be used to liquefy part of the helium, the said coolant gas being helium and each separate stream of coolant gas being expanded through its corresponding turbine from a pressure lower than that of the helium being cooled in the corresponding cooling zone.

9. A method as claimed in claim 8, wherein the cooled helium is expanded and thus further cooled to liquefy part of the helium, and an additional final cooling stage is included prior to the expansion and liquefaction step, in which final cooling stage the coolant is unliquefied helium returned from the liquefaction step.

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