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S. KIESSKALT ET AL

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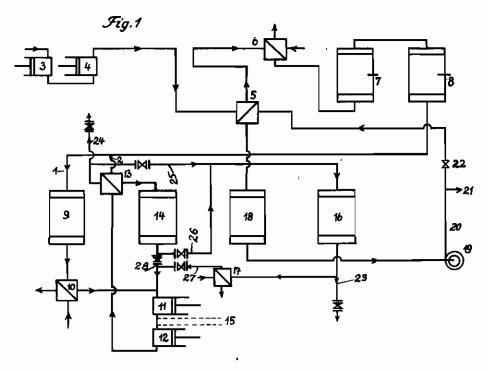
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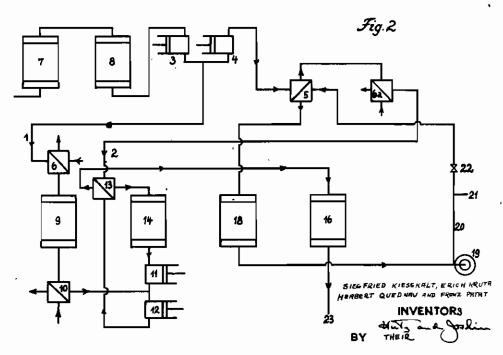
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2 Sheets-Sheet 1





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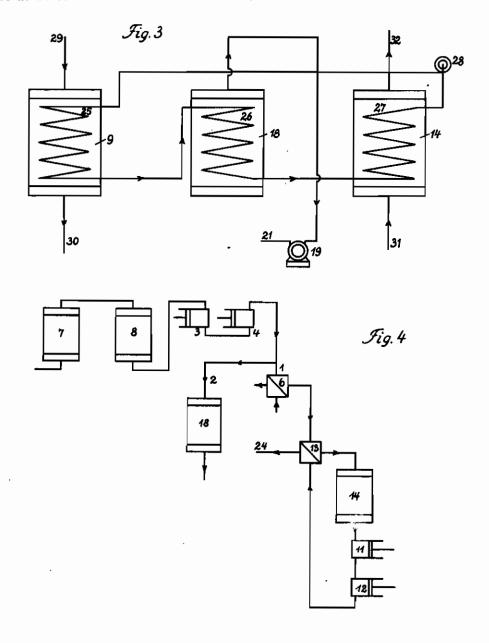
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SIEGFRIED KIESSKALT, ERKH KRUTA, HERBERT QUEDNAU AND FRANZ PATRI INVENTORS

BY THEIR

ATTORNEYS

ALIEN PROPERTY CUSTODIAN

PROCESS OF ADSORBING GASES AND VAPORS

Siegfried Kiesskalt, Erich Kruta, Herbert Quednau, and Franz Patat, Frankfurt-am-Main, Germany; vested in the Allen Property Custodian

Application filed January 3, 1941

The co-pending U.S. Patent Application Serial No. 360,291 filed on October 8, 1940, in the name of Siegfried Klesskalt, relates to a method of obtaining valuable gaseous or liquid substances from a mixture of gases by cooling the mixture under 5 pressure within a cold gas machine and applying adsorbing agents. According to said invention the adsorbers applied are coupled with a cold gas system. Cold gas machines have hitherto not been applied with success in practice, but 10 according to the process described in the aforenamed application a surprising effect is attained by very high additional adsorptions even if a large portion of gas has a low percentage of the material to be obtained. The process, therefore, 15 operates in an economical manner. By the displacement of the adsorption in the pressure sphere with a cooling process simultaneously produced therewith of the entire portion of gas, adsorption isotherms are attained which within 20 the operating sphere still have a substantially steep and straight-lined course, i. e. there exists an optimum of the cooling and of the increase in pressure; the adsorption process thus operates in the most economical manner.

On further investigating the afore-named process we have now found that it may be suitable to subdivide the adsorption process into several stages. The subdivision may relate to the temperature as well as to the pressure. In this con- 30 nection not only thermo-dynamic advantages are produced which, however, are not very essential and, as regards the economical utilization of the machines, do not exceed some per cents, as experience has shown, but in the refrigerating 35 economy simplifications and a saving of far more than 30 per cent of the expenditure of energy have been attained by the new subdivision process.

The invention is diagrammatically illustrated 40 by way of example in the accompanying drawings 1 to 4.

In Fig. 1 there is illustrated, by way of example, how the process may be carried out when the adsorption process is performed in one pressure 45 cycle, but when it is subdivided into two different stages of temperature. The example relates to a process of separating ethylene from illuminating gas. Said process consists in first compressing the entire volume of gas in com- 50 pressors 3 and 4, removing the heat of compression by a heat exchanger 5 and a cooling device 6 and eliminating the benzene by means of an active carbon-plant 7 with application of pressure at ordinary temperature. The active carbon- 55 order to cool the current of gas 2. Before the

plant 7 may also be replaced by a washing oilplant and may likewise be used suitably with application of pressure and at ordinary temperature. The gas is then dried in a dryer 8. After having passed the drying plant 8 the cycle of gas is subdivided into two currents, the proportion of which is destined in the first place by the heat of adsorption which is set free in the various stages.

The partial current I is caused to enter, while applying the increased pressure chosen and a temperature which approximately corresponds to the temperature of the cooling water, an adsorber 9 which contains active carbon and acts as a preliminary adsorbing device. In said adsorber 9 the hydrocarbons to be adsorbed are removed from the current in correspondence with the operating pressure and an average temperature which increases, in correspondence with the heat of adsorption, above the temperature of the cooling water; the adsorbing operation is continued until the adsorbing means are saturated, i. e. until no more material to be gained is adsorbed. The heated current of gas leaves the 25 adsorber 9, is passed to a cooling device 10 where It is allowed to cool until it has reached the temperature of the cooling water and is then caused to flow into expansion devices 11 and 12 which, according to the size of the plant and the pressure chosen, may be constructed as a piston engine, a rotary machine or a turbo machine, in one stage or in several stages.

The partial current 2 which has left the drying plant is cooled in the cold exchanger 13 at the operating pressure to a temperature below ordinary temperature according to the process described in the afore-named application. It is then led to a further adsorber 14 which in a previous operation has been preliminarily charged at a laised pressure and at ordinary temperature in the same manner as the adsorber 9 and now acts as a subsequent adsorbing device. The adsorber 14 is, therefore, now additionally charged; said charge is in correspondence with the difference of the adsorption isotherms at the emperature of the cooling water and at the temperature below ordinary temperature to be attainable.

When current 2 has left the adsorber 14 it is combined with current 1, the combined current acts on the expansion devices 11 and 12, while delivering energy to the compressor plant and cooling the total quantity of gas. Part of the cold thus produced may be led off at 15, or the cold is totally carried to the cold exchanger 13 in 2 373,012

current of gas 2, after having passed the cold exchanger 13, is delivered to the supply area it may be used for cooling the adsorber 16 which previously has been heated for attaining desorption.

At the same time there is thus attained that the adsorbing means in the adsorber 16 are charged with constituents of the illuminating gas which are finally driven out by passing therethrough a supply of fresh gas. Only a slight heat since the adsorption of heat of the ethylene is substantially compensated by the desorption cold of the gas supplanted.

The use of the residual gas freed from the subcharging the adsorber 16 may, of course, also be carried through in a cycle with application of pressure before the current enters the expansion machines 11 and 12, as it is illustrated by the broken lines in Fig. 1. A water-cooling device 20 17 is in that case inserted between adsorber 19 and expansion devices it and 12. By this arrangement there is attained that the gas substantially freed from the substance which is intended to be obtained is used for charging the 25 adsorbing means whereby the heat of adsorption is removed by cooling water. In the present ease illuminating gas from which ethylene has been removed cools the adsorber 16 and simultaneously methane is adsorbed thereof. The adsorption 30 heat is taken off by the cooling device 17. If said adsorber 18 is inserted again into the cycle of the fresh gas still containing the substance which is to be obtained, the methane is removed from the adsorbing means and the ethylene is adsorbed, 35 but a particular heat tone is not produced. The process, therefore, involves the advantage that cold is produced by the total quantity of gas; said cold has, however, to cool only part of the current, for instance half of the gas introduced. 40 In comparison with the process described in the afore-named application there is attained by the additional improvement of the present invention that, though the same supply of energy is used, smaller devices are employed which are much more charged.

The desorption of the ethylene is shown in a particular cycle wherein the ethylene desorbed and conducted through a tube 20 to the heat exchanger 5. The heat taken up in the heat exchanger 5 is used for further desorption in the adsorber 18. If enough ethylene has been desorbed and the pressure in the afore-said cycle 55 exceeds a certain measure, part of the ethylene is removed at 21. After the desorption has been finished, the valve 22 is closed and the pump 19 removes all of the sthylene at 21.

The illuminating gas freed from ethylene is led 60 to the supply area at 23 or, if the cycle on the broken lines is chosen, at 24. The desorption as shown here for adsorber 18 is performed alternatively with the other adsorbers.

A further improvement in comparison with the 65 afore-named application is attainable by a subdivision into several stages of the pressure sphere, as it diagrammatically illustrated in Fig. 2. Said subdivision resides in the following: After having eliminated benzene by plant I and having caused 70 the gas to pass through the dryer 8 there is operated for instance in the first stage after the compression in the compressor 3 under a pressure of 3 atmospheres above atmospheric pressure and at the temperature of the cooling water (current 1); 75 current 2, i. e. into a smaller total portion of gas.

only half the current of gas is branched off and is brought by the compressor 4 from 3 to 8 atmospheres above atmospheric pressure (current 2), so that a subsequent adsorption takes place not only at a lower temperature, but also at a raised pressure, when compared with the previous adsorption stage.

The further process steps shown in Fig. 2 correspond with the steps shown in Fig. 1; the refertone sets in during said exchanging operation 10 ences are the same as in Fig. 1. It may also be advisable to subdivide the process into three or four stages instead of two stages.

The high additional charge of the adsorbing agent which charge has been rendered possible. stance which was to be obtained for cooling and 15 as already mentioned, to a surprising extent by the application of the known cold gas machines. allows of making use of the isothermal discharge known as such by decrease of pressure. After having taken out the fully charged adsorber 18 by expansion to normal pressure, in some cases, and this has been found to be still more advisable by a subsequent evacuation, it is possible rapidly and readily to remove the very high additional charge from the active carbon. If a strong cooling occurs in the discharge adsorber 18, by which cooling a delay would be caused, a compensation cycle may suitably be provided by the three adsorbers, i. e. the preliminary adsorber 9, the additional adsorber 14 and the discharge adsorber 18. by coils or other interchanging elements. Said cycle likewise allows an improvement of the energy economy and an improved utilization of the apparatus. As illustrated in Fig. 3, the coils 25, 26 and 27 inserted in the three adsorbers 9, 18 and 14 are mounted in the cycle in such a manner that the carrier liquid, suitably methanol which will not freeze, is conducted into adsorber 18 which is to be discharged. By the carrier liquid heat is to be transferred from adsorber 9 to adsorber 18 wherein the liquid is cooled. For completing the cycle the methanol then passes the coils of the additional adsorber 14; the decrease of the temperature in the adsorber 18 in comparison to adsorber 14 is thus again rendered utilizessentially lower temperatures may be applied and 45 able. The carrier liquid is moved by a pump 28. Arrow 29 demonstrates the inlet, arrow 30 the outlet of the gas of the adsorber 9, arrow 31 the inlet and arrow 32 the outlet of gas of the adsorber 14; 19 is a pump for exhausting the material to is removed from the adsorber 18 by the pump 19 50 be gained from the adsorber 18 to the gasometer 21.

> Furthermore it is possible, if a two-stage subdivision of the expansion machine is applied, to remove the cold between the two stages, as shown in Fig. 1 at 15; said cold may be used for a subsequent liquefaction of gases.

In cases where no complete separation, but only an enrichment of the material to be obtained, for instance ethylene, has to be attained, the heat required for the desorption can be taken from the heat of compression. The compressed mixture of gases is subdivided, as illustrated in Fig. 4, into two parts. After the benzene has been removed at 7 and the fresh gas has been dried at 8, the latter is compressed by compressors 3 and 4. The current of gas is then subdivided. The heat of compression of current I is removed by passing it through a water cooling device 8. After having passed the cold exchanger 13 current 1 is conducted over active carbon in the adsorber 14 and the ethylene is eliminated. Later on the ethylene is desorbed in the same adsorber by the hot current 2 as shown in adsorber 18. The starting portion of ethylene is thus transferred into the

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Current I free from ethylene is suitably expanded at II and I2 in the same manner as shown in Fig. 1. The cooling occurring thereby may be utilized in the cold exchanger 13.

The method of operation herein described may 5 be used with advantage in the various pressure stages of a plant for the liquefaction of gas. There is thus attained that the higher and economically more expensive pressure stages have to overcome only a fraction of the initial portion of 10 gas. This is of particular importance if a large quantity of waste gas is carried along which cannot or must not be liquefied.

If already from the very beginning compressed gas is present for use, for instance a gas from a long distance gas supply and if it is intended to operate with a gas enriched in the substance intended to be obtained or with a mixture of gases under atmospheric pressure it is suitable to desorb with a gas under atmospheric pressure. For this purpose current 2 is expanded and allowed to pass through a cold exchanger wherein it gives off the expansion cold to current 1 or to a cooling liquid which is conducted through an adsorber.

as possible into the steep, lined branches of the adsorbing its, therefore, no fundament present invention if an adaptive active carbon or silica gel.

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After the adsorber has been charged by current 1, current 2 is used for desorbing. Current 2 is thus enriched in the substance to be obtained. The expansion cold of current 1 may also be used 5 for a higher additional charge if the volume of the apparatus is decreased. It is, of course, also possible to perform the same process by starting with gas which is under normal pressure. In that case only current 1 is compressed, whereas 10 current 2 is directly used for desorption.

By all the processes herein described it is intended by a suitable selection of pressure and temperature, to displace the adsorption as much as possible into the steep, substantially straightlined branches of the adsorption iso-therms. It is, therefore, no fundamental modification of the present invention if an adsorber through which a liquid, for instance a washing oil, is caused to trickle is used instead of an adsorber filled with sertive carbon or silica gel

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