

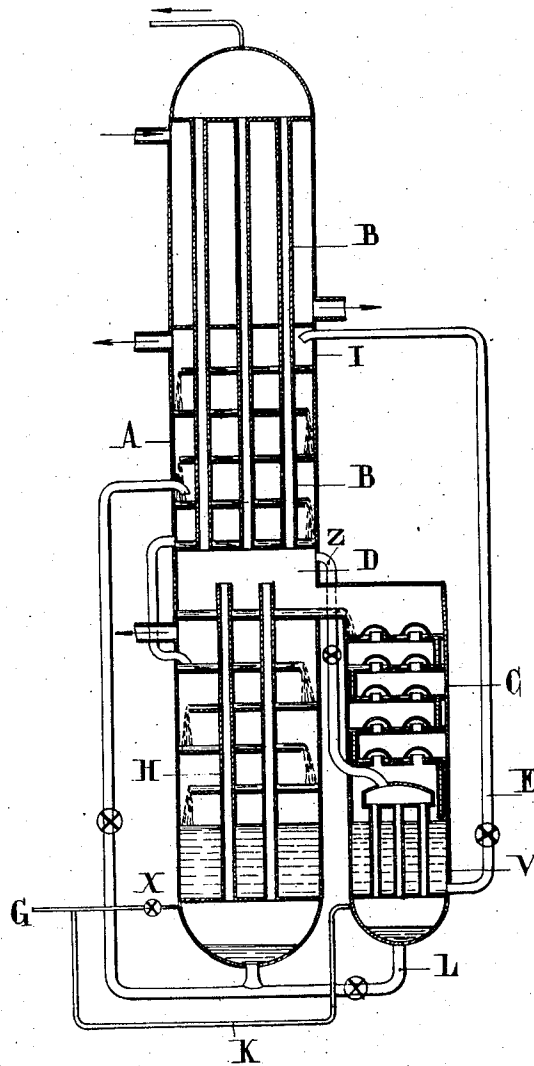
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G. CLAUDE

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GAS SEPARATION PROCESS

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UNITED STATES PATENT OFFICE

GEORGES CLAUDE, OF PARIS, FRANCE, ASSIGNOR TO SOCIÉTÉ L'AIR LIQUIDE, SOCIÉTÉ ANONYME POUR L'ÉTUDE ET L'EXPLOITATION DES PROCÉDÉS GEORGES CLAUDE, OF PARIS, FRANCE

GAS-SEPARATION PROCESS

Application filed May 8, 1925, Serial No. 28,940, and in France June 3, 1924.

This invention relates to the manufacture of hydrogen by the partial liquefaction of gaseous mixtures containing hydrogen and nitrogen, such as coke oven gases, ordinary illuminating gas or the like.

It has been observed that owing to the nitrogen contained in these gases being the most difficultly liquefiable of the constituents other than hydrogen, the liquid nitrogen which is produced towards the end of the progressive liquefaction to which these compressed gases are subjected constitutes a means for effectively eliminating the carbon monoxide ordinarily contained in the said gases, provided that a sufficient quantity of the nitrogen be present. It may happen however that this is not the case, since the liquid produced in the upper nest of tubes of the apparatus heretofore used and discharged outside the said nest of tubes, may contain a considerable proportion of nitrogen which is altogether lost so far as concerns the washing of the carbon monoxide and also the production of ammonia, if the gaseous mixture produced is intended to be used.

The object of the present invention is to avoid such loss of nitrogen as above referred to and thus to improve the efficiency of the process with regard to the purity of the hydrogen obtained thereby. For this purpose according to the present invention the process for the manufacture of hydrogen by partial liquefaction of mixtures of gases containing the same is characterized in that the liquid containing nitrogen produced during the course of the partial liquefaction is subjected in an auxiliary rectification column to a progressive rectification or vaporization whilst under the same pressure as that at which the partial liquefaction of the gases is effected, the gas rich in nitrogen thus obtained returning to the place where the nitrogen containing liquid is produced and the liquid substantially free from nitrogen produced as a result of the said progressive rectification being used for bringing about by its vaporization the said partial liquefaction.

In order that the said invention may be clearly understood and readily carried into effect, the same will now be described more

fully with reference to the accompanying drawing which shows diagrammatically, by way of example, an apparatus suitable for carrying out the present process.

The column A in which the progressive partial liquefaction of the gases is effected is provided with the upper nest of tubes B for the final liquefaction beneath and to one side of which is located a rectifying column C in open communication with the collector D arranged at the bottom of the nest of tubes B. At the lower part of the column C there is provided a vaporizer V connected with which is a pipe E.

The compressed gaseous mixture to be treated, after having been previously cooled to a low temperature, is delivered to the common inlet pipe G; the greater portion of the gases ascends through the lower nest of tubes H in the column A, whence the gases pass into the upper nest of tubes B. The liquid formed in these latter tubes B, containing a certain proportion of nitrogen, falls into the collector D, and thence descends over the successive plates of the rectification column C; during this descent it becomes rectified by contact with the ascending vapors and finally reaches the vaporizer V entirely or almost entirely freed from nitrogen; the liquid from the vaporizer V is discharged through the pipe E into the compartment I outside the nest of tubes B. The heating of the liquid in the vaporizer at the bottom of the column C is effected by a part of the compressed gases to be treated which are delivered through the pipe K from the common inlet pipe G. The gaseous residue from the liquefaction of the compressed gases which pass through the nest of tubes in the vaporizer V is returned to the principal column A, for example by passage through the pipe Z into the collector D. The liquid formed by the liquefaction which takes place inside the nest of tubes of the vaporizer V, being only a small quantity, can be allowed to pass through the pipe L and thereby mix with the liquid which collects at the bottom of the column A. By regulating the valve X the gases to be treated can if desired be admitted to the tubes in the vaporizer V at a pressure slightly

higher than that of the gases which enter the tubes H in the column A.

I claim as my invention:—

1. A process for the manufacture of hydrogen by partial liquefaction of mixtures of gases containing the same, such as coke oven gas or ordinary illuminating gas, in which the liquid containing nitrogen produced during the course of partial liquefaction is subjected in an auxiliary rectification column to a progressive rectification or vaporization whilst under the same pressure as that at which the partial liquefaction of the gases is effected, the gas rich in nitrogen thus obtained returning to the place where the nitrogen containing liquid is produced and the liquid substantially free from nitrogen produced as a result of the said progressive rectification being used for bringing about by its vaporization the said partial liquefaction.
2. A process according to claim 1, in which the liquid in the vaporizer at the bottom of the auxiliary rectification column is heated by passing part of the compressed gases to be treated by indirect contact with said liquid in the said vaporizer whereby the said compressed gases are partially liquefied, the liquid produced as a result of the partial liquefaction being added to that which collects at the bottom of the principal liquefying column whilst the gaseous residue from the aforesaid partial liquefaction is delivered at an appropriate point to the principal liquefying column for further treatment therein.
3. A process according to claim 1, in which the liquid in the vaporizer at the bottom of the auxiliary rectification column is heated by circulating in indirect contact therewith part of the compressed gases to be treated, at a pressure higher than that of the main part of the gases to be treated in the principal liquefying column.
4. A process of manufacturing hydrogen, comprising the steps of partially liquefying a gaseous mixture containing hydrogen; extracting the nitrogen-containing liquid formed during the liquefaction operation; subjecting it to a progressive vaporization out of the path of the gaseous mixture undergoing liquefaction; and mixing the gas rich in nitrogen obtained from the vaporization operation with the gaseous mixture at the point where the nitrogen-containing liquid is collected.
5. A process of manufacturing hydrogen, comprising the steps of partially liquefying a gaseous mixture containing hydrogen in a main liquefying column; delivering to an auxiliary liquefying column, for progressive vaporization therein, the nitrogen-containing liquid formed and collecting in the main column during the liquefaction operation; returning from the auxiliary column to the portion of the main column wherein said nitrogen-containing liquid collects, the gas rich

in nitrogen obtained from the vaporizing operation; and returning the substantially nitrogen-free liquid, which collects in said auxiliary column as a result of the progressive vaporizing operation, to the upper part of the main column so as to travel through the same in counter-current with the hydrogen-containing gaseous mixture and, by its vaporization, effect the partial liquefaction of said mixture.

6. A process of separating the constituents of gaseous mixtures, such as those containing hydrogen, by partial liquefaction, which comprises progressively liquefying the gaseous mixture; collecting the liquid containing the last liquefied constituent; separately subjecting it to a progressive vaporization out of the path of the gaseous mixture undergoing liquefaction; and mixing the gas obtained by said vaporization with the gaseous mixture while undergoing liquefaction.

7. A process of separating the constituents of gaseous mixtures, such as those containing hydrogen, by partial liquefaction, which comprises progressively liquefying the gaseous mixture; collecting the liquid containing the last liquefied constituent; separately subjecting it to a progressive vaporization; returning the gas obtained by said vaporization to that part of the liquefaction process where the last liquefiable constituent liquefies; and vaporizing the liquid remaining after said vaporization for assisting in the liquefaction process.

8. A process of separating the constituents of gaseous mixtures, such as those containing hydrogen, by partial liquefaction, which comprises progressively liquefying the gaseous mixture; collecting the liquid containing the last liquefied constituent; separately subjecting it to a progressive vaporization out of the path of the gaseous mixture undergoing liquefaction; mixing the gas obtained by said vaporization with the gaseous mixture while undergoing liquefaction and utilizing a part of the gaseous mixture to be treated for effecting the progressive vaporization.

9. A process of separating the constituents of gaseous mixtures, such as those containing hydrogen, by partial liquefaction, which comprises progressively liquefying the gaseous mixture; collecting the liquid containing the last liquefied constituent; separately subjecting it to a progressive vaporization; returning the gas obtained by said vaporization to that part of the liquefaction process where the last liquefiable constituent liquefies; and partially vaporizing the liquid remaining after said vaporization by partially liquefying a part of the gaseous mixture treated.

10. A process of separating the constituents of gaseous mixtures, such as those containing hydrogen, by partial liquefaction, which comprises progressively liquefying the gaseous mixture; collecting the liquid

containing the last liquefied constituent; separately subjecting it to a progressive vaporization; returning the gas obtained by said vaporization to that part of the liquefaction process where the last liquefiable constituent liquefies; partially vaporizing the liquid remaining after said vaporization by partially liquefying a part of the gaseous mixture treated; and joining the gas remaining from such partial liquefaction to the main gaseous mixture under liquefaction.

In testimony whereof I affix my signature.

GEORGES CLAUDE.