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COMPLETE SPECIFICATION

Improvements in or relating to the Purification of Gases

We, HENRI MARTIN GUINOT, a citizen of the French Republic, of 31 Rue de Vergennes, Versailles, Seine-&-Oise, France, and SOCIETE POUR L'EQUIPEMENT DES INDUSTRIES CHIMIQUES, S.P.E.I.C.H.I.M.-Reunion des Anciens Etablissements Barbet, Egrot & Grange, Pingris & Mollet-Fontaine, a body corporate organised under the laws of France, of 14 rue La Boetie, Paris, France, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to the purification of gases and more particularly to the separation of hydrogen sulphide and, if desired, carbon dioxide from gaseous mixtures containing the same.

Industrial gases, more especially gases obtained in cracking processes (i.e. produced in oil-refining operations), and natural petroleum gases, almost always contain, among other impurities, carbon dioxide and hydrogen sulphide, sometimes in considerable quantities, which it is necessary to remove before the gases can be used.

It has already been proposed to eliminate carbon dioxide from gaseous mixtures by washing with aqueous solutions of salts capable of giving rise to a reversible reaction, such as the following:

$$AM + CO_2 + H_2O \rightleftharpoons MNCO_3 + AH$$

where M represents a metal, A is an anion and AH represents the acid corresponding to the anion A.

During the washing of the gas, which is carried out in the cold and preferably under pressure, the reaction takes place from left to right, i.e. bicarbonate is formed and the acid AH is liberated.

In the second phase, which also takes place in the cold, but in an inert atmosphere and without pressure, the reverse reaction takes place, i.e. the acid AH acts on the bicarbonate transitorily formed, giving rise to an evolution of carbon dioxide, which is separated. The salt AM is reconstituted without any heating being required. In this way the initial conditions are finally restored so that the cycle of operations may be repeated.

The salts preferably employed in the aforesaid process are alkali salts such as chromates, disodium or dipotassium phosphate, molybdates and, in general, any salt of appropriate alkalinity and capable of giving, under the action of carbon dioxide, an acid compound capable of subsequently decomposing the bicarbonate which is transitorily formed.

It has now been discovered that disodium hydrogen phosphate Na_2HPO_4 and dipotassium hydrogen phosphate K_2HPO_4, which are capable of transitorily fixing carbon dioxide gas, as just explained, are also capable of absorbing increased quantities of hydrogen sulphide under similar operating conditions.

However, hydrogen sulphide is an acid of substantially lower strength than carbon dioxide gas. This is also why only solutions having a strongly basic reaction, such as caustic alkali solutions, ammoniacal liquors, ethanolamines, trisodium phosphate and double alkali metal ammonium phosphates such as sodium diammonium phosphate have hitherto been used to absorb the hydrogen sulphide.

It appears that the action of a dipotassium hydrogen phosphate solution on hydrogen sulphide proceeds according to the equation:

$$K_2HPO_4 + H_2S \rightleftharpoons KSDH + KH_2PO_4.$$

According to the present invention, therefore, a process for separating hydrogen sulphide from gases containing the same and possibly also containing carbon dioxide, comprises washing the gases with an aqueous solution of disodium hydrogen phosphate or dipotassium hydrogen phosphate and thereafter reducing the pressure on the wash liquor to cause liberation of the hydrogen sulphide, and possibly carbon dioxide, and thus regenerate the initial washing solution.

The washing treatment is preferably carried out under superatmospheric pressure using any method industrially known for promoting liquid/gas contact.

Preferably the reduction in pressure is effected by application of a slight vacuum and while the wash liquor is maintained in agitation and under an inert atmosphere. More particularly, it is advantageous to effect a scavenging with inert gas, such as nitrogen, hydrogen, methane or carbon monoxide, for example by injecting the gas through the solution.

In practice, the Na_2HPO_4 or K_2HPO_4 solution may be continuously circulated between a station where it is brought into contact with the gas to be purified and there absorbs the hydrogen sulphide and possibly carbon dioxide, and a station for desorbing the hydrogen sulphide and any carbon dioxide which has been transitorily combined, where, as indicated above, the salt solution is regenerated.

The following data will enable the mechanism of the invention to be readily understood:

A stream of hydrogen sulphide is passed under reflux through an aqueous K_2HPO_4 solution containing 1.5 g. molecules of salt, i.e. 261 g. per litre, at normal temperature and pressure. A quantity of gas immediately passes into solution and it is titrated by iodometry. The dissolved gas is then driven off by vigorously agitating the solution while injecting into it a small quantity of inert gas such as air or nitrogen. When the inert gas ceases to entrain hydrogen sulphide, the liquor is again titrated by iodometry. The absorption and desorption of H_2S is then repeated several times in order to determine whether the composition of the liquor remains constant or not in the course of these operations. The following are the results obtained with a succession of six cycles of absorption and desorption of hydrogen sulphide by the dipotassium hydrogen phosphate liquor.

	H_2S absorbed per litre of solution at 0°C.	H_2S remaining after degassing
1st cycle	6.950 litres per litre	0.840 litre pre litre
2nd cycle	6.850 ,, ,, ,,	0.300 ,, ,, ,,
3rd cycle	6.600 ,, ,, ,,	0.400 ,, ,, ,,
4th cycle	6.900 ,, ,, ,,	0.300 ,, ,, ,,
5th cycle	6.600 ,, ,, ,,	0.300 ,, ,, ,,
6th cycle	6.980 ,, ,, ,,	0.300 ,, ,, ,,

It will be seen from these experiments that the effectiveness of the salt solution in fixing H_2S is maintained constant and that it is capable of holding about 2.2 times as much H_2S as can pure water (the solubility of H_2S in water at 15°C. is 2.945 litres per litre).

The quantity of H_2S which can be fixed per unit volume of liquid naturally varies with the salt concentration and the partial pressure in the gas to be treated.

The following figures show the effectiveness of the process of purification in the case of a washing solution containing 1.5 g. molecule of K_2HPO_4 per litre.

Partial pressure of H_2S in gas	Litres of H_2S absorbed at 20°C. per litre of solution
0.14 atm.	2.5 litres
0.5 atm.	4.8 litres
1 atm.	6.6 litres
4 atm.	12.4 litres
8 atm.	15.9 litres

When it is desired to eliminate simultaneously the carbon dioxide gas and the hydrogen sulphide contained in the same gas, the purification may be effected by means of a single washing provided that the volume of liquor employed is sufficient to fix the whole of whichever of the two impurities is more difficult to eliminate in the case under consideration.

WHAT WE CLAIM IS:—

1. A process for separating hydrogen sulphide from gases containing the same and possibly also containing carbon dioxide which comprises washing the gases with an aqueous solution of disodium hydrogen phosphate or dipotassium hydrogen phosphate, and thereafter reducing the pressure on the wash liquor to cause liberation of the hydrogen sulphide and possibly carbon dioxide and thus regenerate the initial washing solution.

2. A process according to claim 1, wherein the washing treatment is carried out under superatmospheric pressure.

3. A process according to claim 1 or 2, wherein the reduction of pressure is effected by application of a slight vacuum.

4. A process according to any of claims 1 to 3, wherein the reduction of pressure is effected while the wash liquor is maintained in agitation and under an inert atmosphere.

5. A process according to any of claims 1 to 4, wherein the washing liquor is continuously circulated between a station at which it is brought into contact with the gas and a station at which it is subjected to reduction of pressure for the removal of the absorbed gas.

6. A process for separating hydrogen sulphide and, if desired, also carbon dioxide, from gases containing the same, substantially as hereinbefore described.

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