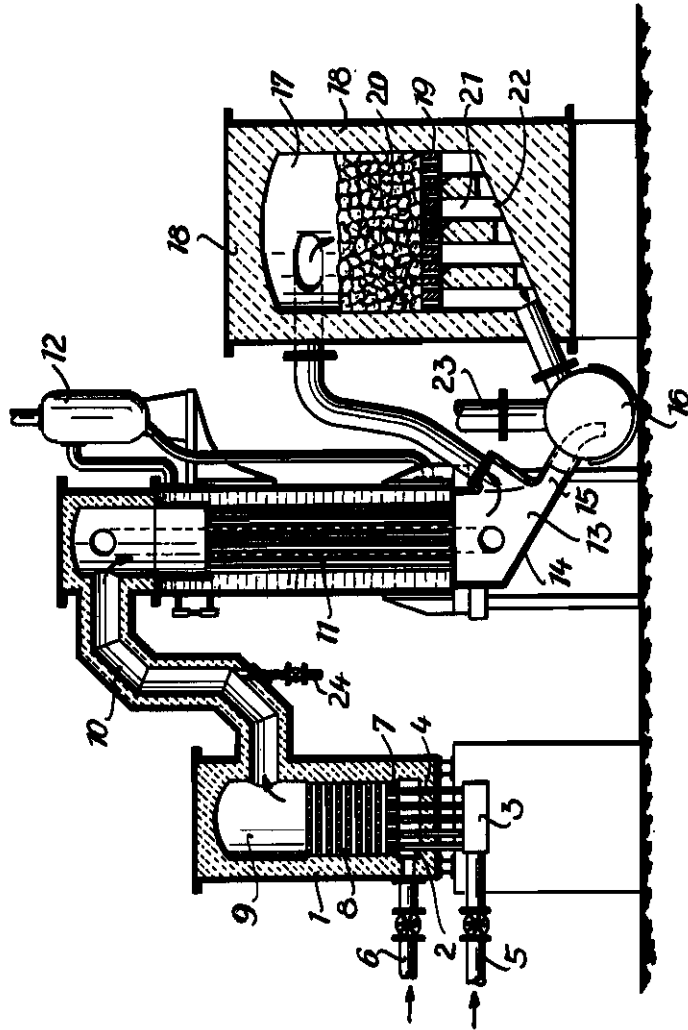


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RECOVERY OF SULPHUR FROM HYDROGEN SULFIDE
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RECOVERY OF SULPHUR FROM HYDROGEN SULFIDE OR HYDROGEN SULFIDE CONTAINING GASES

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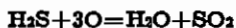
Application filed October 23, 1940

The invention relates to the recovery of elementary sulphur from hydrogen sulphide or hydrogen sulphide containing gases by acting upon the gases with oxygen (air) preferably in the presence of catalysts as bauxite and more particularly to the recovery of sulphur from such gases which contain beside hydrogen sulphide cyanogen compounds or other reactive nitrogen compounds or both of them.

It is a known method to recover elementary sulphur from hydrogen sulphide in that the hydrogen sulphide is mixed with a definite quantity of oxygen (air) and is converted according to the following reaction equation:



Beside this reaction certain side reactions take however place between the hydrogen sulphide and oxygen, for instance according to the equation:



It has been found that the recovery of sulphur from gases containing hydrogen sulphide is rendered difficult or even impossible due to these side reactions if the gases contain also compounds of cyanogen or other nitrogen compounds as amines, ammonia and the like. The sulphur cannot be separated from these gases in such a way that it collects in form of a regulus. The sulphur precipitates rather in more or less large drops which are very viscous and do not join at all or only very slowly. It also happens that this viscous sulphur deposits on the bodies serving as catalysts as for instance on the bauxite pieces and sticks to them so that the surface of the catalyst is covered with a dense coat of sulphur and becomes ineffective.

The inventor now found out that said difficulties in the treatment of gases which contain beside hydrogen sulphide other compounds of cyanogen or other nitrogen compounds arise because the ammonia which is either contained in the gases to be treated or is formed by the partial decomposition of cyanogen compounds, amines or the like, converts with sulphur dioxide resulting from said side reaction and with water into salts of ammonia of the sulphurous acid. These salts of ammonia are solid at the temperatures in question and mix in form of more or less fine crystals with the fluid sulphur so that its viscosity is badly influenced.

Now, the object of my invention is to improve the recovery of sulphur from gases which contain beside hydrogen sulphide cyanogen compounds or other nitrogen compounds in such a

manner that the recovered sulphur collects as a regulus and may be easily removed from the reaction chamber.

According to the invention the gas to be treated is acted upon in an insulated chamber of sufficient size with that quantity of oxygen (air) which is required for the formation of sulphur at such temperatures that the reactive nitrogen compounds contained in the gas beside hydrogen sulphide are decomposed.

It is easily possible to recover the sulphur in a pure, fluid form from the gas resulting from this reaction by cooling the reaction media. According to this invention it is further possible to bring the gaseous and vaporous media formed in the said reaction chamber, into contact with suitable catalysts for the purpose of increasing the yield of sulphur, the temperature of the reaction substances being reduced either before or during the treatment with the catalysts.

Finally it is also possible according to the invention to effect the conversion of the gases to be treated with oxygen (air) in a chamber insulated against heat losses, in which chamber suitable catalysts are provided for which support the conversion between hydrogen sulphide and oxygen as for instance catalysts containing chromeoxide.

In order to decompose the troublesome nitrogen compounds in the gases to be treated it may become necessary to add a larger quantity of oxygen (air) than is required for the conversion of the hydrogen sulphide in elementary sulphur. In case the quantity of oxygen (air) is unfavourably high it is possible according to the invention to add a certain amount of reducing gases as for instance hydrogen or hydrogen containing gases, e. g. purified coal distillation gas to the media formed in the reaction chamber.

With the above and other objects and features of my present invention in view, I shall now describe a preferred embodiment thereof on the lines of the accompanying drawing in which a contrivance for carrying out the invention is shown partly in side view and partly in a vertical section.

The reaction chamber in which the gases to be treated are converted with oxygen (air) is formed by the refractory brickwork 1 which is favourably surrounded by a heat insulating material. At the bottom of the chamber a series of pipes 2 is provided which extend from a distributing chamber 3 and have a certain distance from one another. The pipes 2 traverse the chamber bottom 4.

The hydrogen sulphide containing gas to be treated may be introduced into chamber 3 through pipe 5 in a regulable quantity. The gas is distributed uniformly through the pipes 2 over the whole cross section of the reaction chamber.

The oxygen or the oxygen containing gas (air) required for the conversion is introduced through pipe 6 in a regulable manner. The pipe 6 opens into the intermediate spaces between the pipes 2. The air meets the hydrogen sulphide containing gas introduced through pipes 2, in a layer 7 consisting of refractory bodies arranged above the upper part of the pipes 2 within the reaction chamber. Above the layer 7 furthermore a filling 8 of suitable bodies is provided for which improve the conversion between hydrogen sulphide and oxygen.

According to the invention the filling 8 preferably consists of pipe-like bodies which are made in accordance with the method generally used for the manufacture of refractory bodies by mixing, shaping and burning. These shaped bodies may contain chrome oxide or another substance which improves the reaction between hydrogen sulphide and oxygen or the bricks may be coated with such a substance.

If the contrivance is set into operation first of all the filling 8 of the reaction chamber is brought to the desired high temperature, for instance by burning a suitable fuel gas. If after that the hydrogen sulphide mixed with the required quantity of oxygen (air) is introduced into the chamber to replace the heating gas, the reaction proceeds in the filling 8 all by itself because the required high temperature is maintained by the exothermic reaction between hydrogen sulphide and oxygen.

The gases passed through the filling 8 reach then the free space 9 above the filling. They may be drawn off by means of pipe 10. The gases passing into pipe 10 contain already a considerable quantity of elementary sulphur. Generally 75% of hydrogen sulphide contained in the original gas have been converted into sulphur which may be recovered in fluid form from the gas by means of a simple cooling.

For the cooling of the reaction media a vertical boiler 11 is provided for which may be connected to a vapour collecting vessel 12. But also every other suitable cooling device may be applied in which the gases and vapours are indirectly cooled and the separated fluid sulphur may run off the cooling faces.

At the contrivance shown on the drawing, the fluid sulphur separated by cooling the reaction media collects in chamber 13 below the boiler. The bottom 14 of the collecting chamber 13 is inclined so that the sulphur flows off to the tank 16 through pipe 15.

The reaction gases and vapours remaining in the boiler 11 after the separation of sulphur still contain a considerable amount of sulphur or sulphur compounds. For the recovery of this sulphur a catalyst chamber 17 is provided for at the contrivance shown on the drawing. This chamber 17 is built of refractory brickwork 18. It has a grate-like bottom 19 on which a layer of catalysts is arranged. The catalyst suitably consists of bauxite or another substance which accelerates the conversion of hydrogen sulphide with oxygen to sulphur at comparatively low temperatures.

Below the grate 19 a tank 21 is provided for in which the fluid sulphur leaving the catalyst layer collects. The sulphur flows off the inclined bottom of tank 21 to the collecting tank 16.

The gaseous and vaporous constituents still

existant after the conversion are drawn off the collecting tank 16 through pipe 23 and may be passed through dust chambers, scrubbers or similar contrivances in which the residual injurious sulphur compounds or sulphur dust are removed from the gases. The rest gases escape through a chimney.

The reaction chamber 1 is designed in such a way that it may be brought to such a temperature that the whole nitrogen compounds contained in the gases to be treated are decomposed. The temperature which can be reached depends on the content of hydrogen sulphide in the gases, on the size of the reaction chamber and its heat insulation. If the hydrogen sulphide content of the gas is too low, the gas to be treated is preheated in a suitable manner before it is mixed with air as is common practice at the heating of industry furnaces.

The temperature of the reaction chamber should preferably range at least at 900° C. It is, however, advisable to keep it higher and temperatures of 1200-1300 degrees Centigrade have proved very advantageous for the decomposition of the injurious nitrogen compounds into inert constituents.

In the cooler or boiler 11 connected to the reaction chamber 1 the temperature of the reaction media is reduced to such a degree that the elementary sulphur separates in fluid form. The sulphur may for instance settle out at 130-140° C.

If after the removal of sulphur the residual gases shall undergo a further treatment for the recovery of sulphur it is more advantageous to cool the reaction media from the reaction chamber 1 only to a temperature of about 250° C and after that to pass them on at this temperature to the device 17 which is connected to the cooler. The known reaction between hydrogen sulphide and oxygen is only considerably accelerated by catalysts as bauxite at temperatures above 210° C. It would therefore be of little use to bring the gases into contact with the catalyst below this temperature range.

If, however, the temperature of the reaction media is very much reduced in the cooler 11 to let us say 130° C in order to separate the sulphur in fluid form as far as possible in the cooler and the collecting tank connected therewith the gases are heated up again before entering the catalyst device 17 in a suitable way, so that they have a temperature of 220° C or more within the device 17.

Instead of arranging the catalysts in a special device as shown on the drawing at 17 it is also possible to provide them within the cooler or boiler 11. This may be favourable because the heat which is liberated by the conversion of the hydrogen sulphide with oxygen at the catalysts is immediately led off so that the catalysts cannot be superheated. The catalysts are in this case inserted in the pipes of the cooler or boiler and are kept there by suitable means as for instance by perforated sheet plates or the like.

If the addition of oxygen (air) to the gases to be treated is higher than necessary for the conversion of the hydrogen sulphide into sulphur and for the decomposition of the injurious nitrogen compounds, the yield of elementary sulphur may be considerably reduced as more sulphur dioxide is formed. In order to attain in this case the highest yield of sulphur possible, the invention provides to add reducing gases and preferably those gases which contain hydrogen to the reaction gases which leave chamber 1. Purified

coal distillation gas may be advantageously used, i. e. a gas which is free of injurious sulphur and nitrogen compounds. If the reducing gas still contains hydrocarbons it is advisable to decompose the latter before the gas is added to the reaction media. Such decomposition may be easily arrived at by treating the gas with a moderate quantity of oxygen (air) at increased temperature whereby the hydrocarbons are decomposed and burnt respectively.

At the contrivance shown on the drawing, the reducing gases may be introduced in regulable

quantities through pipe 24 into pipe 10 which leads from the reaction chamber 1 to the cooler 11.

In the process according to the invention the injurious nitrogen compounds are decomposed up to the formation of inert constituents which cannot react with the sulphur or sulphur compounds any more. The products which result from the decomposition of the nitrogen compounds are therefore essentially nitrogen, carbon dioxide and water.

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