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(54) Title: PROCESS FOR PRODUCING POWER FROM A SOUR GAS

(57) Abstract: A process for producing power from a sour gas comprising natural gas and hydrogen sulphide, the process comprising the steps of: (a) combusting the sour gas comprising natural gas and H2S in a combustion-heat recovery device using an excess amount of a oxygen containing gas, resulting in a hot effluent gas comprising carbon dioxide and sulfur dioxide; (b) sending at least part of the hot effluent gas to a heat recovery steam generator to generate steam at least partly used to generate power and a cooled gas effluent comprising C02 and S02; and (c) leading at least part of the cooled gas effluent comprising C02 and S02 to a sulphuric acid unit to produce sulphuric acid, steam and a cleaned flue gas stream.

PROCESS FOR PRODUCING POWER FROM A SOUR GAS

The present invention relates to a process for producing power from a sour gas comprising H2S. The method is particularly useful when combined with a sulfuric acid unit.

Sour gas comprising H2S can originate from various sources. For example, numerous natural gas wells produce sour natural gas, i.e. natural gas comprising H2S and optionally other contaminants. Natural gas is a general term that is applied to mixtures of light hydrocarbons and optionally other gases (nitrogen, carbon dioxide, helium) derived from natural gas wells. The main component of natural gas is methane. Further, often other hydrocarbons such as ethane, propane, butane or higher hydrocarbons are present.

Sales gas production from highly sour gas is expensive, e.g., due to the high costs and complexity of the gas treating facilities to remove all contaminants, including all sulphur species. Often, the produced sales gas is used for power production.

It is desirable to reduce the amount of hydrogen sulphide in sour gas for a number of reasons. Sulphur-containing compounds, such as hydrogen sulphide and oxides of sulphur, are controlled by emission standards in many countries. Furthermore, especially hydrogen sulphide and its combustion products can cause corrosion of equipment.

Normally, sour gases are treated in a so-called acid gas removal unit and the resulting acid gas is sent to a Claus process to produce elemental sulphur. The resulting sweet gas or sweetened gas, comprising the main part of the hydrocarbons, can then be utilized to produce power.

Hydrogen sulphide is recovered from the acid gas stream. This hydrogen sulphide rich gas is then subjected to a multi-step process, e.g., the Claus process, which produces sulphur from gaseous hydrogen sulphide.

The Claus process comprises two stages, a first thermal stage and a second catalytic stage. In the first thermal stage, a portion of the hydrogen-sulphide in the gas is oxidised at temperatures above 850 °C to produce sulphur dioxide and water:

$$2\ H_2S + 3\ O_2 \rightarrow 2\ SO_2 + 2\ H_2O \qquad (I)$$

In the second catalytic stage, the sulphur dioxide produced in the thermal step reacts with hydrogen sulphide to produce sulphur and water:

$$2\ SO_2 + 4\ H_2S \rightarrow 6\ S + 4\ H_2O \qquad (II)$$

The gaseous elemental sulphur produced in stage (II) can be recovered in a condenser, initially as liquid sulphur before further cooling to provide solid elemental sulphur. In some cases, the second catalytic step and sulphur condensing step can be repeated more than once, typically up to three times to improve the recovery of elemental sulphur and reduce emissions of sulphur species.

The second catalytic stage of the Claus process requires sulphur dioxide, one of the products of reaction (I). However, hydrogen sulphide is also required. Typically approximately one third of the hydrogen sulphide gas is oxidised to sulphur dioxide in reaction (I), in order to obtain the desired 1:2 molar ratio of sulphur dioxide to hydrogen sulphide for reaction to produce sulphur in the catalytic stage (reaction (II)). The residual off-gases from the Claus process may contain combustible components and sulphur-containing compounds, for instance when there is an excess or deficiency of

oxygen (and resultant overproduction or underproduction of sulphur dioxide). Such combustible components can be further processed, suitably in a Claus off-gas treating unit, for instance in a Shell Claus Off-gas Treating (SCOT) unit.

The overall reaction for the Claus process can therefore be written as:

$$2\ H_2S + O_2 \rightarrow 2\ S + 2\ H_2O \qquad (III)$$

As conventional Claus installations are costly, both in terms of capital expenditure as well as in terms of operational costs, alternative processes have been reported.

In WO-A-2011134847 a process is described wherein power is produced from a sour gas comprising H2S, wherein in a first step the H2S present is being separated from the natural gas, followed by separate combustion of the cleaned natural gas in a gas turbine and separate burning of the H2S rich stream to also recover energy from the H2S present.

A disadvantage of the process described in WO-A-2011134847 is that two separate burners are being used to generate power. Another disadvantage is the need for an upstream gas treating process that captures the hydrogen sulphide from the sour gas. Due to the high pressure and corrosive nature of sour gas and the complexity of sulfur species present in the gas, this upstream treating generally has a high complexity and incurs high capital expenditure and operating costs. This is especially the case if the sour gas comprises many other contaminants, including sulphur containing contaminants, besides H2S.

It is an object of the invention to provide a process for generating power more efficiently from a sour gas comprising hydrogen sulphide.

It is a further object of the invention to provide a process wherein the generation of power from a sour gas is combined with the production of sulphuric acid.

It is a further object of the invention to provide a process wherein the generation of power from a sour gas is performed whilst reducing the complexity of the process.

To this end, the invention provides a process for producing power from a sour gas comprising natural gas and hydrogen sulphide, the process comprising the steps of: (a) combusting the sour gas comprising natural gas and H2S in a combustion-heat recovery device using an excess amount of an oxygen containing gas, resulting in a hot effluent gas comprising carbon dioxide and sulfur dioxide; (b) sending at least part of the hot effluent gas to a heat recovery steam generator to generate steam, at least partly used to generate power, and a cooled gas effluent comprising CO2 and SO2; and (c) leading at least part of the cooled gas effluent comprising CO2 and SO2 to a sulphuric acid unit to produce sulphuric acid, steam and a cleaned flue gas stream.

The process according to the invention uses the chemical energy present in highly sour natural gases more efficiently to produce power. The invention is suitable for sour gases wherein the sour gas comprises preferably in the range of from 1 to 50 vol% H2S, more preferably in the range of from 10 to 35 vol% H2S. In addition the invention allows to produce sulphuric acid by leaving out the production of sulphur.

By burning the whole sour gas stream the number of process steps and the related process equipment is reduced and at the end better use of the chemical energy present in highly sour natural gases has taken place.

This is especially the case if the sour gas comprises many other contaminants, including sulphur containing contaminants, besides H2S. These contaminants, like mercaptans, require extra treating steps to remove them from the sour gas to produce a cleaned natural gas suitable to be burned in a conventional gas turbine.

However, the sour gas cannot be easily burned. According to prior art processes a sour gas is treated to generate a cleaned natural gas stream for power generation in a so-called combined cycle involving a gas turbine and a steam turbine. For burning of the sour gas including all the H2S and other sulphur species present, the normal combined cycle involving a gas turbine and a steam turbine would not suffice due to the incompatibility of the materials used in the turbine with hydrogen sulphide and its combustion products.

In the process of our invention, care needs to be taken that sufficient oxygen is provided to the process to ensure complete combustion of the feed stream and to avoid that formed SO2 can react with the H2S present according to the Claus reaction, whereby elemental sulphur is being formed, which may condense or even solidify the process equipment. To this end, the oxygen containing gas stream is added to the sour feed gas stream in such amount that a super stoechiometric amount or excess amount of oxygen relative to all combustible components (i.e., hydrocarbons, H2S, mercaptans, COS, CS2, elemental sulfur, BTEX) in the sour gas feed stream is obtained. After the oxidizing process has been completed, the excess amount of the oxygen containing gas is in the range of from 0.1 to 25 vol%, preferably in the range of from 0.1 to 20 vol%, more preferably in the range of from 0.1 to 15 vol%, even more preferably in the

range of from 0.5 to 5 vol%, and even more preferably in the range of from 1 to 2 vol%, this volume% based on the resulting hot effluent gas. By using such excess amount of oxygen, the formation of solid sulphur is being prevented. The formation of solid sulphur may cause fouling to the equipment.

The oxygen containing gas used to combust the sour gas in step (a) of the process is air, oxygen enriched air or oxygen. In order to omit the need to separate air to provide oxygen-enriched air or pure oxygen it is preferred to use air to combust the sour gas stream.

In the combustion process in step (a) part of the SO2 formed may be further oxidized to SO3. Preferably, less than 5% of the SO2 formed is further oxidized to SO3, more preferably less than 3%, even more preferably in the range of from 0.5 to 1.5% of the SO2 formed is further oxidized to SO3.

According to a preferred embodiment, the invention provides a process for producing power from a sour gas comprising natural gas and hydrogen sulphide, the process comprising the steps of: (a) combusting the sour gas comprising natural gas and H2S in a burner-boiler configuration using an excess amount of a oxygen containing gas, resulting in a hot effluent gas comprising carbon dioxide and sulfur dioxide; (b) sending at least part of the hot effluent gas to a heat recovery steam generator to generate steam used, at least partly used to generate power, and a cooled gas effluent comprising CO2 and SO2; and subjecting at least part of the cooled gas effluent comprising CO2 and SO2 to an SO2 concentration step, thereby generating a concentrated gas stream comprising at least 70% SO2 on dry basis and a first cleaned flue gas; (c) leading the concentrated gas

stream comprising at least 70% SO2 on dry basis to a sulfuric acid unit to produce sulfuric acid, steam and a second flue gas stream.

In case SO3 has been formed by oxidation of SO2 in the burner, this SO3 is preferably removed from the effluent stream prior to feed the gas to the SO2 concentration step. The advantage of removing SO3 prior to the SO2 concentration step is a reduced make-up requirement for the SO2 solvent. Solvent make-up leads to increased water and chemicals consumption and emissions. The solvent make-up may be a liquid treating process, for example an ion exchange process.

The sour gas might contain liquids and solid particles when it is coming from the well. It might be useful to first remove such liquids and solid particles, before the sour gas is being combusted in the boiler configuration. Preferably, the liquids and solid particles are being removed by the use of knock-out drums or filter elements, before step (a) of the process. This enhances the life-time of the boiler configuration, because for example plugging of small channels in the burner and erosion of turbine blades is prevented. The sour gas might contain a significant portion of heavier hydrocarbons, so-called condensate and so-called natural gas liquids. To extract the value of these components it may be decided to extract these components prior to combustion, e.g., in a hydrocarbon dewpointing process.

The combustion-heat recovery device is preferably a burner-boiler configuration or a gas turbine - heat recovery combined cycle, also called a combined cycle gas turbine (CCGT). In the preferred situation that the combustion-heat recovery device is a burner-boiler configuration, the burner-boiler configuration comprises

a burner where the combustion takes place, and the heat present in the formed flue gas is transferred to produce steam in a boiler. This boiler can be operated to produce subcritical steam or supercritical steam or ultrasupercritical steam. Preferably, supercritical steam is being produced, as this has the advantage of higher efficiency in the generation of power, without the need for very exotic construction materials as needed with for example ultrasupercritical steam. The steam formed is at least partly used to drive one or more steam turbines.

The steam turbines may be selected from the group consisting of: backpressure turbines, condensing turbines, backpressure/condensing turbines, condensing/extracting turbines, condensing/admission turbines, and condensing/extraction/admission turbines. In another embodiment, the steam turbines may be used to drive one or more of the group consisting of electrical generators, pumps and compressors.

The stack temperature downstream the steam turbines or the heat recovery steam generation (HRSG) unit is, under normal conditions, kept as low as possible to obtain the maximum amount of generated power, and is determined by the minimum amount of draft required in the stack to be able to vent the flue gas. According to the invention, the stack temperature is preferably between 150°C and 300°C, more preferably between 180°C and 280°C, depending on exact fuel composition. The advantage of such higher stack temperatures is that no condensation of corrosive components in the heat recovery steam generation occurs which allows the use of standard HRSG units and no specially designed corrosion resistant materials are required.

In another embodiment, heat may be cogenerated with power by extracting steam from the steam turbines. The steam may be extracted at a pressure of 5 bara, and may be fed to any steam consumer (such as reboilers, live steam injection, general heat exchangers). The extraction pressure level is typically dictated by the anticipated application of the steam. Preferably, steam is extracted to regenerate the solvent that might be used in a SO2 concentration step, to produce a concentrated SO2 stream. To this end the steam can be used to provide heat to the reboiler or provide mechanical power to drive a compressor in a mechanical vapor recompression system.

Sour gas streams are commonly hydrocarbon streams, for instance natural gas streams. Natural gas is comprised substantially of methane, normally greater than 50 mole%, typically greater than 70 mol% methane. Depending on the source, the natural gas may contain varying amounts of hydrocarbons heavier than methane such as ethane, propane, butanes and pentanes as well as some aromatic hydrocarbons. The natural gas may also contain various amounts of hydrogen sulphide. For instance, some natural gas fields contain natural gas having 15-30% hydrogen sulphide by volume. The gas may also contain other non-hydrocarbon impurities such as H2O, N2, CO2 and the like.

The hot effluent gas that is being generated in step (a) has preferably a temperature in the range of from 500 to 800°C.

In step c) at least part of the cooled gas effluent comprising CO2 and SO2 is being sent to a sulfuric acid unit, which removes sulphur dioxide and uses it to generate sulphuric acid. The sulphuric acid unit can produce sulphuric acid from the sulphur dioxide in the

gas effluent in a manner known in the art. For example, the sulphur dioxide can first be oxidised to sulphur trioxide, SO3, with oxygen from an oxygen-comprising stream such as air. A catalyst, such a vanadium (V) oxide catalyst may be present.

The gaseous sulphur trioxide may then be treated with water, to produce sulphuric acid in an exothermic reaction. In order to control the heat evolved, it is preferred to treat the sulphur trioxide with 2-3 wt% water comprising 97-98 wt% sulphuric acid to produce 98-99 wt% concentrated sulphuric acid.

In an alternative embodiment, the sulphur trioxide can be treated with oleum, $H_2S_2O_7$, to form concentrated sulphuric acid. Such processes together with other methods for manufacturing sulphuric acid from sulphur dioxide are well known to the skilled person. The concentrated sulphuric acid can then be added to water to provide aqueous sulphuric acid.

The resulting cooled gas effluent of step (b), comprising SO_2, is a gaseous mixture comprising predominantly sulphur dioxide, nitrogen, carbon dioxide and the excess amount of oxygen. The amount of nitrogen present depends on the source of the oxygen containing gas used to burn the sour gas. In the preferred case that air is being used as oxygen containing gas, next to oxygen a large amount of nitrogen is being introduced that might require removal before entering the sulphuric acid gas unit. Preferably, this gaseous mixture is at least partly separated or partly concentrated to increase the sulphur dioxide content, e.g. by removing the nitrogen and the carbon dioxide. A gas stream is generated comprising at least 70% SO_2 on dry basis.

The advantage of having a sulphur dioxide concentration step before entering the sulfuric acid unit in step (c) is that the size of the sulfuric acid unit can be decreased. Furthermore, by tailoring the composition of the cooled effluent comprising SO2, one becomes more flexible in the choice of the sulfuric acid unit to be used for the production of sulphuric acid. The sulfuric acid unit may comprise a dry sulphuric acid process, or the contact H2SO4 process, or a wet sulphuric acid process, or both next to each other. Preferably, tailoring the composition of the cooled effluent comprising SO2 might be done by combining the gas stream comprising at least 70% SO2 on dry basis with the non-treated part of the cooled gas effluent comprising SO2 before step (c).

The sulphur dioxide can be concentrated by any process know in the art such as for example by using liquid absorption, e.g. the CanSolv process, adsorption, membrane separation or by condensation of the sulphur dioxide. Sulphur dioxide condenses at much higher temperatures, i.e. at approximately -10°C, than for instance nitrogen. Due to the high condensation temperature of sulphur dioxide, the post combustion separation of sulphur dioxide and nitrogen is preferred to the pre combustion separation of oxygen and nitrogen.

A most preferred manner for sulphur dioxide concentration is by contacting the gas effluent comprising sulphur dioxide (i.e. the mixture comprising sulphur dioxide and nitrogen) with an absorbing liquid for sulphur dioxide in a sulphur dioxide absorption zone to selectively transfer sulphur dioxide from the combustion gas effluent to the absorbing liquid to obtain sulphur dioxide-enriched absorbing liquid and

subsequently stripping sulphur dioxide from the sulphur dioxide-enriched absorbing liquid in the regenerator to produce a lean absorbing liquid and the sulphur dioxide-containing gas.

One preferred absorbing liquid for sulphur dioxide comprises at least one substantially water immiscible organic phosphonate diester.

Another preferred absorbing liquid for sulphur dioxide comprises tetraethyleneglycol dimethylether.

Yet another preferred absorbing liquid for sulphur dioxide comprises diamines having a molecular weight of less than 300 in free base form and having a pKa value for the free nitrogen atom of about 3.0 to about 5.5 and containing at least one mole of water for each mole of sulphur dioxide to be absorbed.

Stripping of sulphur dioxide from the sulphur dioxide-enriched absorbing liquid is usually done at elevated temperature. To provide a more energy-efficient process, steam generated in a heat recovery steam generator unit can be used to provide at least part of the heat needed for the stripping of sulphur dioxide from the sulphur dioxide-enriched absorbing liquid. Preferably, steam generated in steps (b) or (d) may be used for the sulphur dioxide concentration.

In a preferred embodiment of the invention, the steam generated in step (b) and (c) are being collected in one steam collecting vessel. The steam can be distributed to various locations in the process, where heat or power is being required.

Depending on the concentration of the H2S of the sour gas and the SO2 concentration step, one might choose the process for the production of sulphuric acid, and whether a SO2 concentration step is needed. Preferably, when the

sour gas comprises in the range of from 1-20 vol%, more preferably in the range of from 10-20 vol%, even more preferably in the range of from 15-20 vol% H2S, the sulfuric acid unit of step (d) comprises a wet sulfuric acid process. In another preferred embodiment, when the sour gas comprises more than 15 vol% H2S, more preferably in the range of from 20 to 50 vol% H2S, even more preferably in the range of from 20-35 vol% H2S, the sulfuric acid unit of step (d) comprises a dry sulfuric acid process.

In the below examples, it is calculated how much power is produced in mega Watt (MW) for the different cases.

Example 1 (comparative)

According to prior art processes, a sour gas stream was first sweetened, followed by conversion of the sweetened gas in a combined cycle with gas turbines to power. To this end, 500 million standard cubic feet per day (MMscfsd) of raw sour well head gas at a pressure of 20 bar was separated from liquids (such as for example water and condensates) and solid content (such as sand) and fed into an acid gas recovery unit to remove H2S and CO2 present in the gas, and, subsequently routed to a glycol dehydration unit and a compression stage to obtain a sweet fuel gas stream at a pressure of 45 bar.

The H2S and CO2 rich separated stream was routed to a Claus unit to convert at least 95% of all sulfur containing components into elemental sulfur, which was obtained as a liquid. The remaining gas stream was further treated in a tail gas treating unit to further reduce the sulfur content before the stream was emitted.

The resulting sweet fuel gas stream was provided as a fuel to a combined cycle gas turbine (CCGT) power plant.

The flue gas obtained upon combustion was virtually free of sulfur and emitted. The total net amount of power exported from this operation was calculated to be 1454 MW.

Example 2

According to the invention, a sour gas was combusted, using a combined cycle gas turbine. To this end, the same 500 MMscfsd raw sour well head gas at a pressure of 20 bar was separated from any liquids (such as for example water and condensate) and solid content (such as sand), followed by dehydration in a glycol dehydration unit, to give a conditioned sour gas fuel stream with 33%(v/v) H2S, 9.5%(v/v) CO2, 55.6%(v/v) CH4, 0.4%(v/v) N2, 1%(v/v) C2 or higher hydrocarbons and 0.5%(v/v) water.

The conditioned sour fuel gas stream was compressed to 45 bar and provided as a fuel to a combined cycle gas turbine (CCGT) power plant. Furthermore, to combust the fuel gas stream, an excess amount of air was added, such that the concentration of oxygen in the flue gas was still 17 vol%. The stack temperature downstream a heat recovery steam generation (HRSG) unit was kept at a temperature of 250C to avoid condensation of corrosive condensates.

The SO2-rich flue gas generated in the CCGT power plant was quenched to a temperature below 45°C in a quench tower and routed through a wet electrostatic precipitator to remove any solids or droplets to give a conditioned flue gas stream with 0.7 vol% SO2.

95 % of the conditioned flue gas stream was fed into an SO2 capture plant, using Cansolv technology. The resulting stream comprised 92.5 vol% SO2. This gas was recombined with the remaining 5 % of conditioned flue gas

stream. The split was chosen such that the combined stream contained 12 vol% SO2. The combined stream was fed into a sulfuric acid plant to produce sulfuric acid.

The total net amount of electrical power exported from this operation was calculated to be 1642 MW.

C L A I M S

1. A process for producing power from a sour gas comprising natural gas and hydrogen sulphide, the process comprising the steps of:

(a) combusting the sour gas comprising natural gas and H2S in a combustion-heat recovery device using an excess amount of a oxygen containing gas, resulting in a hot effluent gas comprising carbon dioxide and sulfur dioxide;

(b) sending at least part of the hot effluent gas to a heat recovery steam generator to generate steam at least partly used to generate power and a cooled gas effluent comprising CO2 and SO2; and

(c) leading at least part of the cooled gas effluent comprising CO2 and SO2 to a sulphuric acid unit to produce sulphuric acid, steam and a cleaned flue gas stream.

2. A process according to claim 1, wherein at least part of the cooled gas effluent comprising CO2 and SO2 of step (b) is subjected to an SO2 concentration step before step (c), thereby generating a concentrated gas stream comprising at least 70% SO2 on dry basis and a second cleaned flue gas.

3. A process according to claim 1 or 2, wherein step (a) and step (b) are being executed in a burner-boiler configuration or a gas turbine-heat recovery combined cycle.

4. A process according to anyone of claims 1-3, wherein the oxygen containing gas is air, oxygen enriched air or oxygen, preferably air.

5. A process according to anyone of claims 1-4, wherein the excess amount of the oxygen containing gas is in the range of from 0.1 to 20 vol%, preferably in the range of from 0.1 to 15 vol%, more preferably in the range of from 0.5 to 5 vol%, this volume% based on the resulting hot effluent gas.

6. A process according to anyone of claims 1-5, wherein the sulfuric acid unit comprises a dry sulphuric acid process.

7. A process according to anyone of claims 1-6, wherein the gas stream comprising at least 70% SO2 on dry basis is combined with a non-treated part of the cooled gas effluent comprising SO2 before step (c).

8. A process according to anyone of claims 1-7, wherein the sour gas comprises in the range of from 1 to 50 vol% H2S.

9. A process according to anyone of claims 1-8, wherein the sour gas comprises in the range of from 10 to 20 vol% H2S and the sulfuric acid unit of step (d) comprises a wet sulfuric acid process.

10. A process according to anyone of claims 1-9, wherein the sour gas comprises in the range of from 20 to 35 vol% H2S and the sulfuric acid unit of step (d) comprises a dry sulfuric acid process.