The invention provides a process for removing one or more gaseous contaminants from a contaminated feed gas stream, the process comprising 1) providing the contaminated feed gas stream, 2) cooling the contaminated feed gas stream to a temperature
at which liquid contaminant is formed as well as a contaminant depleted gaseous phase, and 3) separating the two phases by introducing them into a gas/liquid separator vessel, comprising a gas/liquid inlet at an intermediate level, a liquid outlet arranged below the gas/liquid inlet and a gas outlet arranged above the gas/liquid inlet, in which vessel a normally horizontal coalescer is present above the gas/liquid inlet and over the whole cross-section of the vessel and in which vessel a centrifugal liquid separator is arranged above the coalescer and over the whole cross-section of the vessel, the liquid separator comprising one or more swirl tubes. The contaminated gas stream can be a methane-containing gas stream, especially a natural gas stream, or a gas stream from a gasification or combustion process. The invention further provides a device for carrying out the process as well as the purified gas stream.
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Title: PROCESS FOR REMOVING A GASEOUS CONTAMINANT FROM A CONTAMINATED GAS STREAM

Abstract: The invention provides a method for removing one or more gaseous contaminants from a contaminated feed gas stream, comprising the process of providing the contaminated feed gas stream, 2) cooling the contaminated feed gas stream to a temperature at which liquid contaminant is formed as well as a contaminant depleted gaseous phase, and 3) separating the two phases by introducing them into a gas/liquid separator vessel, comprising a gas/liquid inlet at an intermediate level, a liquid outlet arranged below the gas/liquid inlet, and a gas outlet arranged above the gas/liquid inlet, in which vessel a normally horizontal coalescer is present above the gas/liquid inlet and over the whole cross-section of the vessel and in which vessel a centrifugal liquid separator is arranged above the coalescer and over the whole cross-section of the vessel, the liquid separator comprising one or more swirl tubes. The contaminated gas stream can be a methane-containing gas stream, especially a natural gas stream, or a gas stream from a gasification or combustion process. The invention further provides a device for carrying out the process as well as the purified gas stream.
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PROCESS FOR REMOVING A GASEOUS CONTAMINANT FROM A CONTAMINATED GAS STREAM

The invention relates to a process for removing one or more gaseous contaminants, such as carbon dioxide or hydrogen sulphide, from a contaminated gas stream containing methane, especially a natural gas stream. The invention further relates to a device for carrying out the process and to the purified gas stream obtainable by the process.

The removal of acidic gaseous contaminants, especially carbon dioxide and/or hydrogen sulphide, from methane containing gas streams has been described in several publications.

In WO 2004/070297 a process for removing gaseous contaminants from a natural gas stream is described. In this process, water is removed from the feed gas stream by cooling, the cooling resulting in methane hydrate formation. This step is followed by separation of the hydrates and further cooling of the remaining gas stream, resulting in the formation of solid acidic contaminants. After separation of the solid acidic contaminants a cleaned natural gas stream is obtained. Heating is used to convert the solid contaminants into a slurry or liquid. The slurry or liquid is removed from the separation zone. Usually special measures are necessary to remove also the smallest solid particles from the gas stream.

In WO 2003/062725 a process is described for the removal of freezable species from a natural gas stream by cooling a natural gas stream to form a slurry of solid
acidic contaminants in compressed liquefied natural gas. The solids are separated from the liquid by means of a cyclone. It will be clear that a complete separation of the liquid from the solids is not easily achieved.

In WO 2007/030888 a process is described similar to the process described in WO 2004/070297, followed by washing the cleaned natural gas stream with methanol.

In US 4,533,372 a cryogenic process is described for the removal of carbon dioxide and/or hydrogen sulphide from natural gas by treating the feedstream in a distillation zone and a controlled freezing zone.

There is still a need for a further improved process to remove carbon dioxide and/or hydrogen sulphide from natural gas streams. It is especially desired to design a relatively simple process, not requiring any rotating equipment, not requiring cryogenic distillation processes, not requiring the handling of solid particles or liquid/solid slurries and only having a minimum plot space requirement (footprint).

The process according to the present invention comprises no more than two steps requiring relatively simple equipment only. In the first step the contaminated gas stream containing methane is cooled until liquid contaminants condense, followed by a simple gas/liquid separator, not needing any rotating equipment and not needing any energy for a distillation section.

Thus, the process provides a process for removing one or more gaseous contaminants from a contaminated feed gas stream, the process comprising 1) providing the contaminated feed gas stream, 2) cooling the contaminated feed gas stream to a temperature between -30°C and -80°C, at which temperature at which temperature liquid
contaminant is formed as well as a contaminant depleted
gaseous phase, and 3) separating the two phases by
introducing them into a gas/liquid separator vessel, comprising a gas/liquid inlet at an intermediate level, a liquid outlet arranged below the gas/liquid inlet and a gas outlet arranged above the gas/liquid inlet, in which vessel a normally horizontal coalescer is present above the gas/liquid inlet and over the whole cross-section of the vessel and in which vessel a centrifugal liquid separator is arranged above the coalescer and over the whole cross-section of the vessel, the liquid separator comprising one or more swirl tubes.

The process is a rather easy and uncomplicated process. It actually comprises only a cooling step, followed by a simple gas/liquid separation step, the separation step not involving any moving equipment and not comprising any energy consuming steps. When using a vertical gas/liquid separator vessel, the process can be performed in a device only needing a relatively small plot space.

According to a preferred embodiment, the gas/liquid inlet comprises an admittance with a supply and distribution assembly extending horizontally in the separator vessel. In its most simple form, the inlet is a simple pipe, having a closed end and a number of perforations evenly distributed over the length of the pipe. Optionally, the pipe may have a tapered or conical shape. One or more cross pipes may be present to create a grid system to distribute the gas-liquid mixture more evenly over the cross-section of the vessel. Preferably, the assembly includes a chamber, e.g. a longitudinal box-like structure, connected to the gas inlet and having at least one open vertical side with a grid of guide vanes disposed one behind each other, seen in the direction of the flow. By means of this supply and distribution
assembly, the gas is evenly distributed by the guide vanes over the cross-section of the column, which brings about an additional improvement of the liquid separation in the coalescer/centrifugal separator combination. A further advantage is that the supply and distribution assembly separates from the gas any waves of liquid which may suddenly occur in the gas stream, the separation being effected by the liquid colliding with the guide vanes and falling down inside the column. Suitably, the box structure narrows down in the direction of the flow. After having been distributed by the vanes over the column cross-section, the gas flows up to the coalescer. In a preferred embodiment the longitudinal chamber has two open vertical sides with a grid of guide vanes.

Suitable gas/liquid inlets are described in e.g. GB 1,119,699, US 6,942,720, EP 195,464, US 6,386,520 and US 6,537,458. A suitable, commercially available gas/liquid inlet is a Schoepentoeter inlet device.

There are numerous horizontal coalescers available, especially for vertical columns. A well-known example of a coalescer is a wire mesh. All of these are relatively tenuous (large permeability) and have a relatively large specific (internal) plot space. Their operation is based on drop capture by collision of drops with internal surfaces, followed by drop growth on these surfaces, and finally by removal of the grown drop either by the gas or by gravity.

The horizontal coalescer can have many forms which are known per se and may, for example, consist of a bed of layers of gauze, especially metal or non-metal gauze, e.g. organic polymer gauze, or a layer of vanes or a layer of structured packing. Also unstructured packings can be used and also one or more trays may be present.
All these sorts of coalescers have the advantage of being commercially available and operating efficiently in the column according to the invention. See also Perry’s Chemical Engineers’ Handbook, Sixth edition, especially Chapter 18. See also EP 195464.

The centrifugal liquid separator in one of its most simple forms may comprise a horizontal plate and one or more vertical swirl tubes extending downwardly from the plate, each swirl tube having one or more liquid outlets below the horizontal plate at the upper end of the swirl tube. In another form, the centrifugal liquid separator comprises one or more vertical swirl tubes extending upwardly from the plate, each swirl tube having one or more liquid outlets at the upper end. The plate is provided with a downcomer, preferably a downcomer that extends to the lower end of the separator vessel.

In a preferred embodiment, the centrifugal liquid separator comprises two horizontal trays between which vertical open-ended swirl tubes extend, each from an opening in the lower tray to some distance below a coaxial opening in the upper tray, means for the discharge of secondary gas and of liquid from the space between the trays outside the swirl tubes, and means provided in the lower part of the swirl tubes to impart to the gas/liquid a rotary movement around the vertical axis.

The liquid separator is also preferably provided with vertical tube pieces which project down from the coaxial openings in the upper tray into the swirl tubes and have a smaller diameter than these latter. This arrangement enhances the separation between primary gas on the one hand and secondary gas and liquid on the other.
hand, since these latter cannot get from the swirl tubes into the openings in the upper tray for primary gas.

According to a preferred embodiment, the means for discharging the secondary gas from the space between the trays consist of vertical tubelets through the upper tray, and the means for discharging liquid from the space between the trays consist of one or more vertical discharge pipes which extend from this space to the bottom of the column. This arrangement has the advantage that the secondary gas, after having been separated from liquid in the said space between the trays, is immediately returned to the primary gas, and the liquid is added to the liquid at the bottom of the column after coming from the coalescer, so that the secondary gas and the liquid removed in the centrifugal separator do not require separate treatment.

In order to improve even further the liquid separation in the centrifugal separator, openings are preferably provided in accordance with the invention at the top of the swirl tubes for discharging liquid to the space between the trays outside the swirl tubes. This has the advantage that less secondary gas is carried to the space between the trays. A suitable, commercially available centrifugal separator is a Shell Swirltube deck.

In a preferred embodiment, the separation vessel comprises a second normally horizontal liquid coalescer above the centrifugal liquid separator and over the whole cross-section of the vessel. The second coalescer is arranged to remove liquids from the secondary gas. This has the advantage that any droplets still present in the secondary gas are removed as described hereinabove. Preferably, the second coalescer is a bed of one or more
layers of gauze, especially metal or non-metal gauze, e.g. organic polymer gauze. In another preferred embodiment, the second normally horizontal liquid coalescer is situated above the secondary gas outlets, for instance in the way as described in EP 83811, especially as depicted in Figure 4.

The contaminated gas stream may be any gas stream containing acidic contaminants.

Suitably, the contaminated gas stream is a methane containing gas stream, for instance from natural sources as natural gas, associated gas or coal bed methane or from industrial sources as refinery streams or synthetic sources as Fischer-Tropsch streams or from biological sources as anaerobic waste or manure fermentation. The amount of methane present may vary over a wide range, e.g. from 3 to 90 vol%, preferably 5 and 80 vol% methane, more preferably between 10 and 75 vol%.

The contaminated gas stream may also be a syngas stream comprising carbon dioxide, a flue gas stream or a gas stream obtained after combustion of coal. Syngas is a mixture of carbon monoxide and hydrogen. A syngas stream comprising carbon dioxide is generally obtained after converting part or most of the carbon monoxide to carbon dioxide in a so-called water gas shift reaction.

The acidic contaminants in the feedstream are especially carbon dioxide and hydrogen sulphide, although also carbonyl sulphide (COS), carbon disulphide (CS2), mercaptans, sulphides and aromatic sulphur compounds may be present. Beside acidic contaminants, also inerts may be present, for instance nitrogen and noble gases as argon and helium. The amount of acidic contaminants present in the feed gas may vary over a wide range. The amount of carbon dioxide in the feed gas is suitably
refrigerant, e.g. a propane cycle, an ethane/propane cascade or a mixed refrigerant cycle, optionally in combination with an internal process loop, suitably a carbon dioxide stream (liquid or slurry), a cold contaminant depleted stream or washing fluid or in which the cooling is done by compression, cooling and expansion.

The feed gas stream is cooled to a temperature between -30 and -80 °C, preferably between -40 and -60 °C. At these temperatures liquids will be formed.

The raw feed gas stream may be pre-treated to partially or completely remove of water and optionally some heavy hydrocarbons. This can for instance be done by means of a pre-cooling cycle, against an external cooling loop or a cold internal process stream. Water may also be removed by means of pre-treatment with molecular sieves, e.g. zeolites, or silica gel or alumina oxide or other drying agents. Water may also be removed by means of washing with glycol, MEG, DEG or TEG, or glycerol. The amount of water in the gas feed stream is suitably less than 1 vol%, preferably less than 0.1 vol%, more preferably less than 0.01 vol%.

The feedstream for the process of the invention, optionally pretreated as described above, will suitably have a pressure between 10 and 120 bara, or even up till 160 bara. Especially, the feedstream has a pressure between 15 and 70 bara, preferably between 20 and 50 bara. The feedstream, preferably pretreated, suitably has a temperature between -30 and 150 °C, suitably between -20 and 70 °C, preferably between 0 and 50 °C.

In a preferred embodiment of the invention, the contaminant depleted gaseous phase is further purified,
refrigerant, e.g. a propane cycle, an ethane/propane cascade or a mixed refrigerant cycle, optionally in combination with an internal process loop, suitably a carbon dioxide stream (liquid or slurry), a cold contaminant depleted stream or washing fluid or in which the cooling is done by compression, cooling and expansion.

Suitably the feed gas stream is cooled to a temperature between -30 and -80 °C, especially between -40 and -60 °C. At these temperatures liquids will be formed.

The raw feed gas stream may be pre-treated to partially or completely remove of water and optionally some heavy hydrocarbons. This can for instance be done by means of a pre-cooling cycle, against an external cooling loop or a cold internal process stream. Water may also be removed by means of pre-treatment with molecular sieves, e.g. zeolites, or silica gel or alumina oxide or other drying agents. Water may also be removed by means of washing with glycol, MEG, DEG or TEG, or glycerol. The amount of water in the gas feed stream is suitably less than 1 vol%, preferably less than 0.1 vol%, more preferably less than 0.01 vol%.

The feedstream for the process of the invention, optionally pretreated as described above, will suitably have a pressure between 10 and 120 bara, or even up till 160 bara. Especially, the feedstream has a pressure between 15 and 70 bara, preferably between 20 and 50 bara. The feedstream, preferably pretreated, suitably has a temperature between -30 and 150 °C, suitably between -20 and 70 °C, preferably between 0 and 50 °C.

In a preferred embodiment of the invention, the contaminant depleted gaseous phase is further purified,
e.g. by extraction of remaining acidic components with a chemical solvent, e.g. an aqueous amine solution, especially aqueous ethanolamines, such as DIPA, DMA, MDEA, etc., or with a physical solvent, e.g. cold methanol, DEPG, NMP, etc.

The contaminated gas stream is continuously provided, continuously cooled and continuously separated.

The invention is further explained by Figure 1. In Figure 1 a device is shown for carrying out the process of the present invention. A dry feed stream (water content 20 ppm, 60 bara) flows via pipe 1 to turbo-expander 2. In the expander the temperature drops to -45 °C. The feed gas comprises 40 vol% of carbon dioxide, 2 vol% of ethane and 58 vol% of methane. The resulting gas/liquid mixture is introduced into vertical gas/liquid separator 3 via supply and distribution assembly 4. Most of the liquid will flow down to the lower end of the separator and leave the separator via liquid outlet 5. The gaseous stream comprising larger and smaller droplets will flow upwards via liquid coalescer 6, centrifugal separator 7 and a second liquid coalescer 8 to the top of the separator vessel, and leave the separator vessel via gas outlet 9.
**CLAIMS**

1. A process for removing one or more gaseous contaminants from a contaminated feed gas stream, the process comprising 1) providing the contaminated feed gas stream, 2) cooling the contaminated feed gas stream to a temperature between -30°C and -80°C, at which temperature liquid contaminant is formed as well as a contaminant depleted gaseous phase, and 3) separating the two phases by introducing them into a gas/liquid separator vessel, comprising a gas/liquid inlet at an intermediate level, a liquid outlet arranged below the gas/liquid inlet and a gas outlet arranged above the gas/liquid inlet, in which vessel a normally horizontal coalescer is present above the gas/liquid inlet and over the whole cross-section of the vessel and in which vessel a centrifugal liquid separator is arranged above the coalescer and over the whole cross-section of the vessel, the liquid separator comprising one or more swirl tubes.

2. Process according to claim 1, in which the gas inlet comprises an admittance with a supply and distribution assembly extending horizontally in the separator vessel, the assembly consisting of a longitudinal box-like structure connected to the gas inlet and having at least one open vertical side with a grid of guide vanes disposed one behind each other, seen in the direction of the flow.

3. Process according to claim 1 or 2, in which the horizontal coalescer consists of one or more layers of gauze, especially metal or non-metal gauze, or combination thereof.
4. Process according to claim 1 or 2, in which the horizontal coalescer consists of a layer of vanes.

5. Process according to claim 1 or 2, in which the horizontal coalescer consists of a layer of structured packing.

6. Process according to any of claims 1 to 5, in which the centrifugal liquid separator comprises two horizontal trays between which vertical open-ended swirl tubes extend, each from an opening in the lower tray to some distance below a coaxial opening in the upper tray, means for the discharge of secondary gas and of liquid from the space between the trays outside the swirl tubes, and means provided in the lower part of the swirl tubes to impart to the gas/liquid a rotary movement around the vertical axis, preferably in which the liquid separator is provided with vertical tube pieces projecting down from the coaxial openings in the upper tray into the swirl tubes and having a smaller diameter than these latter, or in which the means for discharging secondary gas from the space between the trays consists of vertical tubelets through the upper tray and in which the means for the discharge of liquid from the space between the trays consists of one or more vertical discharge pipes extending from this space to the bottom of the column.

7. Process according to claim 6, in which in the separation vessel a second normally horizontal liquid coalescer is present above the centrifugal liquid separator and over the whole cross-section of the vessel, wherein the second coalescer is arranged to remove liquids from the secondary gas.

8. Process according to claim 7, in which the second coalescer is a bed of one or more layers of gauze,
especially metal or non-metal gauze, or combinations thereof.

9. A process according to any preceding claim, in which the gaseous contaminants are acidic gaseous contaminants, especially carbon dioxide or hydrogen sulphide, preferably in which the natural gas stream comprises between 5 and 80 vol% of methane, preferably between 10 and 75 vol%, or in which the contaminated gas stream contains between 5 and 55 vol% of hydrogen sulphide, preferably between 10 and 45 vol%.

10. A process according to any of the preceding claims, in which the contaminated gas stream has a temperature between -20 and 150 °C, preferably between -10 and 70 °C, and a pressure between 10 and 150 bara, preferably between 15 and 70 bara.

11. A process according to any of claims 1 to 10, in which the cooling is done by isenthalpic expansion, preferably isenthalpic expansion over an orifice or a valve, especially a Joule-Thomson valve, or in which the cooling is done by nearly isentropic expansion, especially by means of one or more expanders, preferably turbo expanders or laval nozzles.

12. A process according to claim 11, in which the feed gas stream is pre-cooled to a temperature between 15 and -35 °C, preferably between 5 and -20 °C, before expansion.

13. A process according to any of claims 1 to 10, in which the cooling is done by heat exchange against a cold fluidum, especially an external refrigerant, e.g. a propane cycle, an ethane/propane cascade or a mixed refrigerant cycle, or an internal process loop, suitably a carbon dioxide or hydrogen sulphide stream or a cold
methane stream or in which the cooling is done by
compression, cooling and expansion.

14. A process according to any of the preceding claims,
in which the contaminated gas stream is cooled to a
temperature between -30 and -80 °C, preferably between
-40 and -60 °C.