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(54) Benævnelse: **FREMGANGSMÅDE OG REAKTIONSSYSTEM TIL FREMSTILLINGEN AF METHANOL**

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DESCRIPTION

Description

[0001] The present invention relates to a process for the preparation of methanol by catalytic conversion of methanol synthesis gas and a reaction system for carrying out the process.

[0002] More particularly, methanol is by the invention prepared in two reaction units, in which a first unit is operated in once-through mode on fresh synthesis gas optionally admixed with unconverted synthesis gas separated from effluent of a second reaction unit and in which the second reaction unit is operated in a synthesis loop with unconverted synthesis gas optionally admixed with fresh synthesis gas.

[0003] The reaction of carbon oxides and hydrogen to methanol is equilibrium limited and conversion of the synthesis gas to methanol per pass through a methanol catalyst is relatively low, even when using a high reactive synthesis gas.

[0004] Because of the low methanol production yield in a once-through methanol conversion process, the general practice in the art is to recycle unconverted synthesis gas separated from the reaction effluent and dilute the fresh synthesis gas with the recycle gas.

[0005] This typically results in the so-called methanol synthesis loop with one or more reactors connected in series being operated on fresh synthesis gas diluted with either recycled unconverted gas separated in a separator from the reactor effluents or on the reactor effluent containing methanol and unconverted synthesis gas. The recycle ratio (recycle gas: fresh synthesis feed gas) is 1 : 1 up to 7 : 1 in normal practice.

[0006] In methanol process designs with a once-through reaction unit and a reaction unit operated on recycled unconverted synthesis in a methanol synthesis loop, optimal gas compositions can be set to the once-through reaction unit or reaction unit operated on recycled unconverted synthesis by the recycle gas and feed bypass gas. The effluent from the once-through reaction unit is mixed with the effluent from the reaction unit in the synthesis loop. Produced methanol is separated from the mixed effluent after cooling upstream separation unit. Separated unconverted synthesis gas contained in the mixed effluent combined from the reaction units is recycled to the reaction unit in the synthesis loop.

[0007] In order to save equipment, the cooling and separation of the methanol containing effluent gas can be made in a combined cooling train. However, the combined cooling train will have mixed the two different recycle gasses, i.e one from the once through reaction unit and one from the synthesis loop reaction unit. To prevent built up of inert gases, a part of the combined recycled unconverted synthesis gas must be purged from the loop, thereby purging

out more active reactants than necessary.

[0008] For the sake of simplicity, in the following description and claims the "once through reaction unit" is termed "first methanol methanol reaction unit" and the "synthesis loop reaction unit" is termed "second methanol reaction unit"

[0009] The term "methanol catalyst" used in the following description and in the claims refers to any catalyst being active in the conversion of hydrogen, carbon monoxide and carbon dioxide to methanol. Those catalysts are not part of the invention and are extensively disclosed in the patent literature.

[0010] Appropriate methanol catalysts for use in the invention are as an example the known copper-zinc based catalysts.

[0011] The main principle of the invention is, thus, to withdraw a hot purge gas from the effluent of the second methanol reaction unit, prior to the effluent is combined with the effluent from the first reaction unit.

[0012] The hot purge gas will allow combined equipment in cooling train and at the highest content of inert components and the lowest activity, which allows more effective purging without losing too much reactants with only one extra small cooler.

[0013] The term "inerts" refers to components contained in methanol synthesis gas, which are not chemically reactive in the methanol synthesis.

[0014] Accordingly, this invention is a process for the preparation of methanol, comprising the steps of

(a) providing a fresh methanol synthesis gas containing hydrogen, carbon monoxide and carbon dioxide;

(b)) introducing and reacting the fresh methanol synthesis gas stream in a first methanol reaction unit in presence of a methanol catalyst and obtaining a first effluent stream containing methanol and unconverted synthesis gas;

(c) providing a recycle gas stream containing the unconverted methanol synthesis gas contained in the first effluent stream and unconverted methanol synthesis gas from a second methanol reaction unit;

(d) introducing and reacting the recycle gas stream in the second methanol reaction unit in presence of a methanol catalyst;

(e) withdrawing a second effluent stream containing methanol and the unconverted methanol synthesis gas from the second methanol reaction unit;

(f)) combining the first and a part of the second effluent stream;

(g) cooling and separating the combined effluent into a methanol-containing liquid stream and the recycle stream; and

(h) withdrawing the remaining part of the second effluent stream as a purge gas stream,

wherein the remaining part of the second effluent stream is withdrawn as a purge gas stream prior to combining the first and second effluent stream.

[0015] In some applications of the process according to the invention, it will be desirous to adjust the module $M=(H_2-CO_2)/(CO+CO_2)$ of the fresh synthesis gas by addition of hydrogen to the gas. Hydrogen can be recovered from the purge gas and recycled to the process upstream the synthesis gas compressor.

[0016] Thus, in an embodiment of the invention at least a part of hydrogen contained in the purge gas stream is recovered and recycled to step b).

[0017] To provide optimum conditions for the methanol reaction in the first methanol reaction unit, a part of the recycle stream is introduced into the first methanol reaction unit in further an embodiment.

[0018] In still an embodiment, a part of the fresh methanol synthesis gas is introduced into the second methanol reaction unit to provide optimum condition for the methanol synthesis in the second methanol reaction unit.

[0019] The first and second methanol reaction unit can comprise one or more reactors selected from boiling water cooled reactors, gas cooled reactors, quench reactors and adiabatic operated reactors, connected in series and/or in parallel.

[0020] The invention provides furthermore reaction system for use in a process for the preparation of methanol, the system comprises a first and second methanol reaction unit containing each a methanol catalyst;

a process gas stream passageway for introducing a process gas stream of a fresh synthesis gas into the first methanol reaction unit and a circulation passageway for circulating unconverted synthesis gas to the second methanol reaction unit;

a first effluent passageway for withdrawing and passing a first methanol containing effluent stream from the first reaction unit to a mixing point in a second effluent passageway for withdrawing a second methanol containing effluent from the second reaction unit;

separating means arranged downstream the mixing point in the second effluent passageway for separating methanol from the unconverted synthesis gas;

a circulator arranged in the circulation passageway between the separating means arranged upstream the second methanol reaction unit; and

a purge gas line connected to the circulation passageway and arranged upstream the mixing point in the second effluent passageway.

[0021] In an embodiment of the reaction system according to the invention, the purge gas line is connected to hydrogen recovery unit.

[0022] In an embodiment of the invention, a passageway is connected to the hydrogen recovery unit and to the process gas stream passageway for passing hydrogen to the gas stream of a fresh synthesis gas.

[0023] In an embodiment of the invention, the reaction system further comprises a passageway connected to the hydrogen recovery unit and to the process gas stream passageway for passing hydrogen to the gas stream of a fresh synthesis gas.

[0024] In an embodiment of the invention, the reaction system further comprises a split stream passageway for passing a part of the unconverted synthesis gas from the circulation passageway to the process gas stream passageway.

[0025] In an embodiment of the invention, the reaction system further comprises a split stream passageway for passing a part of the process gas stream of a fresh synthesis gas to the circulation passageway.

[0026] The first and second reaction unit in the above embodiments can comprise one or more methanol reactors selected from boiling water cooled reactors, gas cooled reactors, quench reactors and adiabatic operated reactors connected in series and/or in parallel.

Patentkrav

1. Fremgangsmåde til fremstillingen af methanol, omfattende trinnene med

(a) tilvejebringelse af en frisk methanolsyntesegas indeholdende hydrogen, carbonmonoxid og carbondioxid;

(b)) indføring og reaktion af den friske methanolsyntesegasstrøm i en første methanolreaktionsenhed i tilstedeværelse af en methanolkatalysator og opnåelse af en første effluentstrøm indeholdende methanol og ikke-omdannet syntesegas;

(c) tilvejebringelse af en recirkulationsgasstrøm indeholdende den ikke-omdannede methanolsyntesegas indeholdt i den første effluentstrøm og ikke-omdannet methanolsyntesegas fra en anden methanolreaktionsenhed;

(d) indføring og reaktion af recirkuleringsgasstrømmen i den anden methanolreaktionsenhed i tilstedeværelse af en methanolkatalysator;

(e) udtagning af en anden effluentstrøm indeholdende methanol og den ikke-omdannede methanolsyntesegas fra den anden methanolreaktions-enhed;

(f)) kombination af den første og en del af den anden effluentstrøm;

(g) afkøling og adskillelse af den kombinerede effluent i en methanolholdig væskestrøm og recirkulationsstrømmen; og

(h) udtagning af den resterende del af den anden effluentstrøm som en rensegasstrøm,

hvor den resterende del af den anden effluentstrøm udtages som en rensegasstrøm før kombination af den første og anden effluentstrøm.

2. Fremgangsmåden ifølge krav 1, hvor mindst en del af hydrogen indeholdt i rensegasstrømmen genvindes og recirkuleres til trin b).

3. Fremgangsmåden ifølge krav 1 eller 2, hvor en del af recirkulationsstrømmen indføres i den første methanolreaktionsenhed.

4. Fremgangsmåden ifølge et hvilket som helst af kravene 1 til 3, hvor en del af den friske methanolsyntesegas indføres i den anden methanolreaktionsenhed.

5. Fremgangsmåden ifølge et hvilket som helst af kravene 1 til 4, hvor den første og anden methanolreaktionsenhed omfatter en eller flere reaktorer valgt blandt kogendevandkølede reaktorer, gaskølede reaktorer, quenchreaktorer og adiabatisk drevne reaktorer forbundet i serie og/eller parallelt.

6. Reaktionssystem til anvendelse i en fremgangsmåde til fremstilling af methanol, hvilket system omfatter en første og anden methanolreaktionsenhed, der hver indeholder en methanolkatalysator; en procesgasstrømpassage til at indføre en procesgasstrøm af en frisk syntesegas i den første methanolreaktionsenhed og en cirkulationspassage til at cirkulere ikke-omdannet syntesegas til den anden methanolreaktionsenhed;

en første effluentpassage til at udtage og lede en første methanolholdig effluentstrøm fra den første reaktionsenhed til et blandingspunkt i en anden effluentpassage til at udtage en anden methanolholdig effluent fra den anden reaktionsenhed;

adskillelsesorgan anbragt nedstrøms for blandingspunktet i den anden effluentpassage til at adskille methanol fra den ikke-omdannede syntesegas;

en cirkulator anbragt i cirkulationspassagen mellem adskillelsesorganet anbragt opstrøms for den anden methanolreaktionsenhed; og

en rensegasledning forbundet med cirkulationspassagen og anbragt opstrøms for blandingspunktet i den anden effluentpassage.

7. Reaktionssystemet ifølge krav 6, hvor rensegasledningen er forbundet med hydrogengenvindingsenhed.

8. Reaktionssystemet ifølge krav 6, yderligere omfattende en passage forbundet med hydrogengenvindingsenheden og med procesgasstrømpassagen til at lede hydrogen til gasstrømmen af en frisk syntesegas.

9. Reaktionssystemet ifølge et hvilket som helst af kravene 6 til 8, yderligere omfattende en delt strømpassage til at lede en del af den ikke-omdannede syntesegas fra cirkulationspassagen til procesgasstrømpassagen.

10. Reaktionssystemet ifølge et hvilket som helst af kravene 6 til 9, yderligere omfattende en delt strømpassage til at lede en del af procesgasstrømmen af en frisk syntesegas til cirkulationspassagen.

11. Reaktionssystemet ifølge et hvilket som helst af kravene 6 til 10, hvor den første og anden reaktionsenhed omfatter en eller flere methanolreaktorer valgt blandt kogendevandkølede reaktorer, gaskølede reaktorer, quenchreaktorer og adiabatisk drevne reaktorer forbundet i serie og/eller parallelt.