

11) Publication number:

**0 269 854** A2

(12)

### **EUROPEAN PATENT APPLICATION**

21) Application number: 87115768.1

(5) Int. Cl.4: **B01J 8/02** , C01C 1/04 , //C07C29/15

2 Date of filing: 27.10.87

3 Priority: 03.11.86 CH 43335/86

43 Date of publication of application: 08.06.88 Bulletin 88/23

Designated Contracting States:
 AT DE ES FR GB GR IT NL

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- System for reducing energy consumption in multi-unit reactors for heterogeneous synthesis and related reactors.
- The system for reducing energy consumption in multi-unit reactors for heterogeneous synthesis, and more particularly for the synthesis of ammonia, with a single bed in each reaction unit, such as for example the "C.F. Braun". consisting of two cylindrical walls at least partly perforated for the annular delimitation of the catalytic beds; the outer wall (Fe) with diameter (Di) slightly smaller than diameter (Dc) of cartridge (C) has height (Hi), while (H'i) is the height of the internal cylindrical wall (Fi) which in turn has diameter (D'i) much smaller than said (Di) and about equal to diameter (Dt) of the central tube (T) for the outlet of reacted gas (RG). The top of internal cylindrical wall (Fi) is closed by a cover (CO) with distance (D) from the upper edge of the external wall of the basket (Fe), the basket being closed at the bottom by a tapered wall (PR). The catalytic bed is therefore: 1) annular with diameter (Di-D'i) for the whole of its height (Hi-D): 2) substantially a full cylinder along height (D): 3) tapered outside (for example a truncated cone), and internal cylindrical cavity with diameter (Di) for the whole of its height (H1 Hi = Hi"). The reacted gas collects in the area (Q) inside the perforated cylindrical wall (Fi).

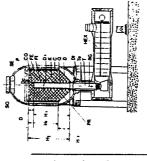


FIG.

# SYSTEM FOR REDUCING ENERGY CONSUMPTION IN MULTI-UNIT REACTORS FOR HETEROGENEOUS SYNTHESIS AND RELATED REACTORS

#### BACKGROUND OF THE INVENTION

Field of the invention

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This invention concerns a system for increasing conversion yields and reducing energy consumption in multiunit reactors for heterogeneous synthesis and more particularly for the synthesis of ammonia, each unit consisting of an outer shell (P), and of an internal cartridge (C) with a diameter (Dc) and total height (H1), and containing a catalyst in granules arranged in a catalytic bed (K) with axial height (H1) and situated between a cylindrical external wall with diameter (Di) and height (Hi) and an inner cylindrical wall with diameter (D'i) (smaller than Di), and height H'i, inside which the gas, the upper traverse face of the bed being open and penetrated axially by the synthesis gas.

It is well known that reactors for catalytic synthesis under pressure, and more particularly for the catalytic synthesis of ammonia, methanol, higher alcohols, etc., consist of an outer shell and of a single unit or of several units according to the C.F. Braun process, each unit having an internal cartridge containing a catalyst in granules in various shapes and with various characteristics arranged in one or more layers (catalytic beds). In reactors according to the C.F. Braun process each unit has a single-layer catalytic bed.

The synthesis gas running through the various catalytic beds is generally cooled between one bed and the next, these being exothermic reactions, in order to achieve optimal temperature conditions in the various catalytic beds by injecting fresh gas (quench reactor) or by direct interchange with the cold incoming gas. Said cooling is carried out in heat exchangers arranged outside and amongst the several units of the reactor where the C.F. Braun process is concerned.

Reactors featuring radial gas flow in the catalytic beds have recently been put forward (Lummus, Topsoe, Kellogg, USA patents No. 3918918 and No. 4181701, European Patent Appl. No. 007743-A1) or featuring axial-radial flow (Ammonia Casale USA Patents No. 4372920 and No. 4405562) which represent a considerable progress when compared to axial flow reactors (including the C.F. Braun reactor), particularly when it is necessary to use large quantities of catalyst, as they reduce pressure drop in the catalytic beds and thus energy consumption. In these instances the axial gas flow requires wide reactors (low length-diameter ratio of the apparatus) with high equipment costs and high energy consumption.

Ammonia Casale USA Patents No. 4372920 and No. 4405562 have made it possible greatly to simplify cartridge design in synthesis reactors, achieving an internal reactor structure easily accessible for maintenance and to load and unload the catalyst with, at the same time, low pressure drop. According to the above mentioned patents, in effect, each catalytic bed consists of a perforated cylindrical external wall, a perforated cylindrical internal wall and only one sealed end (the upper wall of the basket being, in effect, fully open); and upper unperforated portion of said internal cylindrical wall (or of both cylindrical walls), together with the section of open passage situated between the upper edges of the two cylindrical walls arranged on a level approximately perpendicular to the longitudinal axis of said perforated walls, constituting the passage where a minor portion of gas runs through the catalytic bed with a prevalently axial flow, while the remaining major portion of gas runs with radial flow through the major portion of catalytic bed arranged in the perforated area of the two cylindrical walls of the bed.

According to the above invention the portion of gas running through the bed with a prevalently axial flow is controlled by the height of the portion of unperforated internal cylindrical wall (or both internal and external) which in any case according to a prevalent characteristic of the invention is a minor portion of the total height of the wall itself. Reactors with dimensions equal to a height/diameter ratio greater than 10 (i.e., with a diameter/height ratio smaller than 0.1) are preferably to be produced according to the above invention.

In the current economic world situation modernisation of existing plants is becoming increasingly important: the majority of such plants where a synthesis process is concerned (for example the synthesis of ammonia) uses reactors with the gas flowing axially through the catalytic beds (axial reactors) characterised by a low height/diameter ratio of the reactor, owing to the above mentioned need to contain pressure drops in the reactor. In the case of C. F. Braun reactors consisting of several units each with a low height/diameter ratio and each containing a single catalytic bed, the same applies as mentioned above not only for existing reactors but also for the new reactors planned by C. F. Braun with two or more units still according to the above criterion.

In previous Patent Applications, more particularly in European Patent Application No. 86104911.2, the Applicants have already described a system for the reduction of energy consumption in reactors for heterogeneous catalytic synthesis under pressure and more particularly in reactors with dimensions equal to a low height/diameter ratio (less than 10): it is in particular a system suitable for revamping existent axial gas flow reactors, and more particularly Kellogg reactors used in a great number of high capacity plants (800 ÷ 1500 t/d) built towards the end of the Seventies.

In another Patent Application the Applications described a system for revamping "Chemico" axial reactors.

All the above plants using axial reactors are characterised by the high energy consumption not critical in those years when it was available at a low cost, since the axial-type reactor is one of the reasons for high energy consumption.

While continuing their researches the Applicants found a system which can now be easily applied for the modernisation of other axial reactor and more particularly C. F. Braun reactors, whether already in existence or newly built.

The invention, therefore, concerns a system as described in the introductory part of the description and of the main claim, characterised by the fact that the catalytic bed is delimited by:

- a) (inside and in the vicinity fo said outer wall forming cartridge (C) ), by a cylindircal outer wall (Fe) with diameter (Di) permeable by gas for at least the major portion of its height (Hi). (Di) being only slightly smaller than the diameter of cartridge (C):
- b) and internal cylindrical wall (Fi) also permeable by gas for at least the major portion of its height (H'i) with a diameter (D'i) much smaller than the above mentioned (Di) of (Fe) and of the same order of magnitude of diameter (Dt) of the central tube (T) which is the outlet for the reacted gas, the top (SO) of (Fi) being closed by a cover (CO) possible permeable by gas at least in part and with a distance (D) from the top (SE) of the external wall (Fe):
- c) a lower wall (PR) with the height (H"i) along which its diameter (D") decreases towards the bottom. beginning from diameter (Dc) of the cartridge to diameter (Di) of the internal cylindrical wall: the catalytic bed consists therefore of a first solid upper cylindrical area with diameter (Di) and height (D): of a second annular area with diameter (Di-Di) and height (Hi-D): and a third tapered area with height (H"i = H1-Hi) and diameters decreasing from (Di) to (Di).
- The ratio of the gas portions (with a prevalently axial flow and radial flow) will be different in the catalytic beds of the various units where these have a variable height (variable height/diameter ratio of the baskets), as in the application of this invention to modify the above mentioned C.F. Braun reactors, in which the height (Hi) of the internal cartridge's catalytic beds could increase in the various beds in cascade arrangement (2 3 beds each with its own shell)

#### BRIEF DESCRIPTION OF THE DRAWINGS

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The various aspects and advantages of the invention will become more apparent from the following description of the embodiment shown in Fig. 2, opposite the known technique shown in Fig. 1, these illustrations being a schematic view of a longitudinal cross-section. More particularly the following description of a preferred embodiment (modification of the axial-flow C. F. Braun reactor) will better illustrate the features of the invention.

The embodiment described refers (as preference but not as limitation) to the modification of a C. F. Braun ammonia reactor with several catalytic beds (for example three) through which runs the gas with an axial flow, and cooling of the gas outside the beds (reactor shown by way of example in Fig. 1 of the paper "New Synloop Lower Ammonia Production Costs: by Wilson, Grotz & Richez, pages 63 - 79, presented at the British Sulphur Conference in Amsterdam in April 1986).

The reaction gas (see Fig. 1) enters from the bottom of the reactor in 1 and rises in the airspace (I) between shell (P) and cartridge (C), and enters (flow 2) axially from top to bottom the catalytic bed (K). The reacted gas reaches outlet tube (T) which feeds the heat exchanger (HEX) outside shell (P).

Fig. 2 shows the system according to the invention, in which the catalytic bed is now delimited by:

a) (inside and in the vicinity of said outer wall forming cartridge (C) ), by a cylindrical outer wall (Fe) with diameter (Di) permeable by gas for at least the major portion of its height (Hi), (Di) being only slightly smaller than the diameter of cartridge (C);

- b) an internal cylindrical wall (Fi) also permeable by gas for at least the major portion of its height (H'i) with a diameter (D'i) much smaller than the above mentioned (Di) of (Fe) and of the same order of magnitude of diameter (Dt) of the central tube (T) which is the outlet for the reacted gas, the top (SO) of (Fi) being closed by a cover (CO) possibly permeable by gas at least in part and with a distance (D) from the top (SE) of the external wall (Fe):
- c) a lower wall (PR) with height (H"i) along which its diameter (D") decreases towards the bottom, beginning from diameter (Dc) of the cartridge to diameter (D'i) of the internal cylinidrical wall.

The catalytic bed consists therefore of a first solid upper cylindrical area with diameter (Di) and height (D): of a second annular area with diameter (Di-D'i) and height (Hi-D): and of a third tapered area with height (H"i = H1-Hi) and diameters decreasing from (Di) to (D'i).

The following comparative example better illustrates the advantages of the invention.

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### COMPARATIVE EXAMPLE

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# Basic synthesis data by C.F. Braun

25 - Capacity

1360 MTD ammonia

- Pressure at reactor's inlet

210 bar

- Gas composition at reactor's inlet:

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i2 69.00 % mol

N2

23.00 % mol

Ar + CH4

5.00 % mol

инз

3.00 % mol

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## C.F. Braun two-bed reactor

- Diameter 1st bed

about 3000 mm

- Diameter 2nd bed

about 3300 mm

- Catalyst volume (6 ÷ 10 mm) ·

1st bed

20 m3

2nd bed

53 m3

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C. F. Braun two-bed reactor modified with axial-radial beds according to the invention

- Diameter 1st bed : about 3000 mm

- Diameter 2nd bed : about 3300 mm

- Catalyst volume (1.5÷3 mm)

1st bed : 17.4 m3

2nd bed : 50.2 m3

These figures are for a modified reactor already in existence. For new reactors, an optimised L/D ratio would be taken into consideration.

Reactor performance

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C. F. Braun C. F. Braun Conventional with axial-radial internals

- NH3 content at

reactor's outlet 17 % mol 20.6 % mol

- △P reactor --- 2 bar reduction

### CONCLUSIONS

The improvements that can be achieved with axial-radial internals can be summed up as follows:

- -Higher concentration of NH3 at reactor's outlet
- -Reduction of circulation flow (about 20%)
- -Reduction in loop pressure drop (about 35%)
- -Reduction in refrigeration units (about 20%).

The above applies to modified existing reactors. Where new reactors are concerned, the dimensions of catalyst volume (with two or three beds) and of the reactors (L/D ratio) can be optimised with maximum yield and minimal investment, thus achieving even better results.

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#### Claims

- 1. A system for increasing conversion yields and reducing energy consumption in multiunit reactors for heterogeneous synthesis and more particularly for the synthesis of ammonia, each reaction unit consisting of an outer shell (P) and of a cartridge (C) containing a catalyst in granules arranged in a catalytic bed (K) with axial height (H1) and situated between a cylindrical external wall with diameter (Di) and height (Hi), and an inner cylindrical wall with diameter (Di) (smaller than Di) and height (H'i), inside which collects the reacted gas, and a tapered bottom (PR), the upper traverse face of said bed being open and penetrated axially by the synthesis gas, characterised by the fact that the catalytic bed is delimited by:
- a) (inside and in the vicinity of said outer wall forming cartridge (C)), by a-cylindrical outer wall (Fe) with diameter (Di) permeable by gas for at least the major portion of its height (Hi). (Di) being only slightly smaller than the diameter of cartridge (C):
- b) an internal cylindrical wall (Fi) also permeable by gas for at least the major portion of its height (H'i) with a diameter (D'i) much smaller than the above mentioned (Di) of (Fe) and of the same order of magnitude of diameter (Dt) of the central tube (T) which is the outlet for the reacted gas, the top (SO) of (Fi) being closed by a cover (CO) possibly permeable by gas at least in part and with a distance (D) from the top (SE) of the external wall (Fe):
- c) a lower wall (PR) with the height (H"i) along which its diameter (D") decreases towards the bottom starting from the diameter (Dc) of the cartridge towards diameter (D'i) of the internal cylindrical wall.
- the catalytic bed consisting therefore of a solid upper cylindrical area with diameter (Di) and height (D): of a II annular area with diameter (Di-D'i) and height (Hi-D): and a III tapered area with height (H''i = H1-Hi) and diameters decreasing from (Di) to (D'i).
- 2. System in claim 1, characterized by the fact that the reaction gas runs through the I area with a prevalently axial flow, the II area with a prevalently radial flow and the III area with a mixed flow.
- 3. System as in claims 1 and 2, characterized by the fact that within the same reactor the said flows can be adjusted by varying the ratio between heights Hi, H'i, H''i and Di.
- 4. System as in the above claims, characterized by the fact that reactors within the same bank have increasing H1/Di ratios.
  - 5. System substantially as described, exemplified and shown.
- 6. Reactor for heterogeneous synthesis, such as obtained with the system according to the foregoing claims.
  - 7. Reactors substantially as described and shown.

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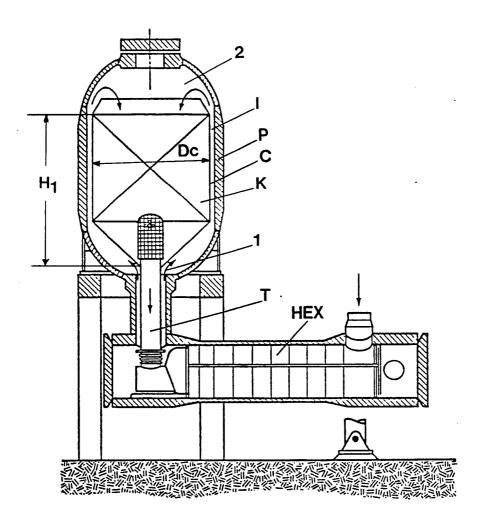


FIG.1

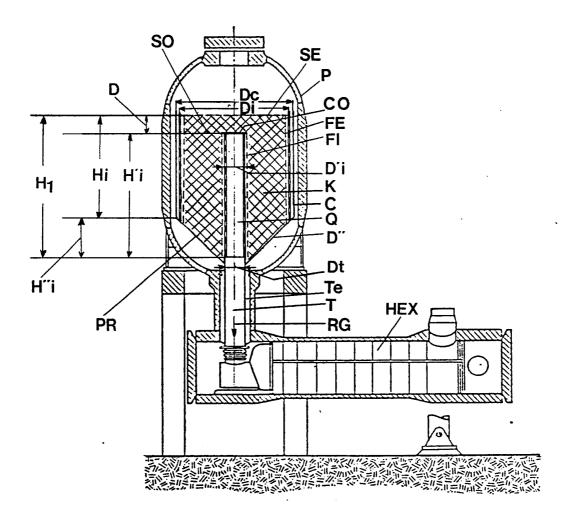


FIG. 2