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(54) Process and reactor for exothermic heterogeneous synthesis with several catalytic beds and with the external recovery of reaction heat.

(57) In a process for exothermic and heterogeneous synthesis, for example of ammonia, in which the synthesis gas is reacted in several catalytic beds (6, 7, 13) with axial-radial or only radial flow, said reaction gas is collected at the outlet from the last catalytic bed but one (7) and is transferred to a system for heat recovery (SAC) external to the reactor, and is re-introduced into the last catalytic bed (13).

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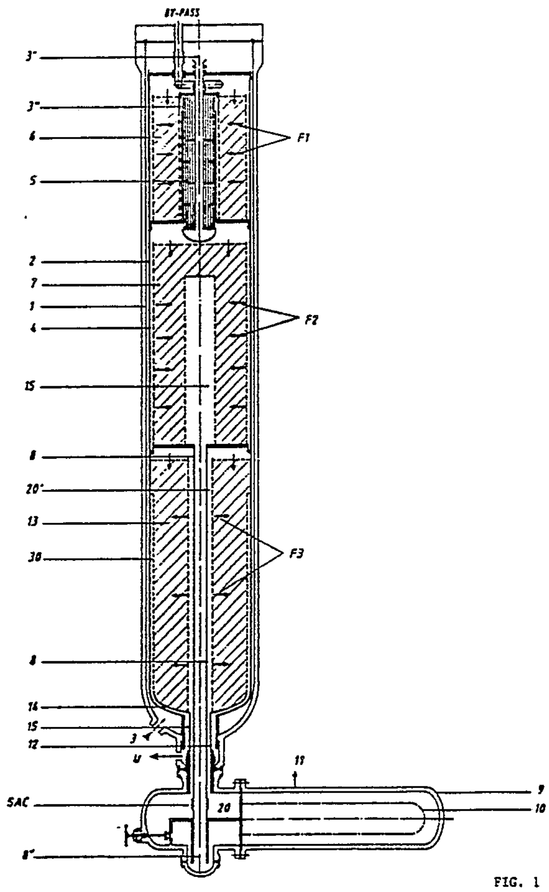


FIG. 1

Process and reactor for exothermic heterogeneous synthesis with several catalytic beds and with the external recovery of reaction heat

This invention concerns a process for exothermic heterogeneous synthesis in which the synthesis gas runs through a series of catalytic beds stacked but separate one from the other contained within the same reaction space, where the gas leaving one bed runs through the next catalytic bed.

The invention also concerns reactors for this process, consisting of a pressure-resistant outer shell, of baskets for catalytic beds all inside the same shell, of a possible cartridgé and of a heat regenerator.

In the production of ammonia the amount of heat which develops from the synthesis reaction of $N2+3H2$ is remarkable, such heat being generally recovered for the final purpose of producing steam used in the production cycle in order to reduce energy consumption.

The latest technologies aim at the maximum recovery of said synthesis heat at the highest degree of heat possible; synthesis units and their principal piece of equipment, the reactor, are therefore suitably designed for this purpose.

In newly built plants the reactors have several catalytic beds with intermediate cooling of the gas by means of indirect exchange through heat exchangers; besides, part of the reaction heat is removed with an external cooling agent such as for example the feed water to the boiler or by generating steam, before the last reaction stage, in order to be able to operate at the maximum temperature possible (heat recovery at the highest degree of heat) without any limitation to the maximum reaction yield achievable.

Maximum temperature together with maximum yield are in effect two contrasting requirements, as is widely demonstrated by the diagrams which show in abscissa the concentration of ammonia and in ordinate the temperature of the gas.

The most important designers of synthesis reactors have generally favoured reactors with several catalytic beds in at least two distinct apparatus units in series, in order to satisfy the above-mentioned need for the optimal exchange of reaction heat (at the highest degree of heat possible) without limiting the meximum yield achievable (Fertilizer Focus, October 1987).

In the case of two separate items or units of equipment, the first of these contains generally two catalytic beds with intermediate indirect cooling by means of an internal exchanger, while the second item or unit of equipment generally contains a single catalytic bed.

Between the two items of equipment heat exchange is achieved by introducing a boiler to produce steam. This is the case for the Topsoe Series 250 (Series 200 1 Series 50) reactor and for the Uhde reactor, both with radial flow of the gas in the catalytic beds (Fertilizer Focus, October 1987, pages 36 and 39).

There are also reactors designed in three separate parts, each containing a catalytic bed with axial gas flow according to the design by C.F. Braun (Nitrogen Conference, Amsterdam 1986). In this case a boiler for the production of steam is introduced between the second and the third reaction unit (Nitrogen Conference, Amsterdam 1986, Mr. K.C. Wilson, Mr. B.J. Grotz and Mr. J. Richez of CdF Chimie).

According to a recent patent by CF Braun (UK Patent Application 2132501A), the gas/gas exchanger between catalytic beds, which is normally conveniently situated inside the reactors with at least two beds within a single unit, is situated outside the reaction unit directly connected at the bottom of the shell containing a single catalytic bed.

To minimize the problem of tubes at a high temperature, the tube connecting the above-mentioned horizontal exchanger with the shell containing the catalytic bed is cooled by the fresh gas fed to the reactor.

After having pre-heated the fresh feed gas, the gas leaving the catalytic bed leaves the exchanger and feeds the unit containing the second ctalytic bed (CF Braun reactor with several reaction units, as shown in Figure 5 of the Wilson, Gritz, Richez report in the above-mentioned reference and on page 48 of Fertilizer Focus, October 1987).

The problem solved in the above-mentioned CF Braun Patent, i.e. the avoidance of high temperature gas coming into contact with the tubes connecting shell and exchanger, does not exist in reactors with several catalytic beds in a single unit since, as mentioned above, the gas/gas exchanger is located directly inside the reactor itself.

Still according to CF Braun the problem of optimal recovery of heat is solved in a complex way by introducing a boiler connected by a complex tube arrangement to the reactor itself (see Figure 5 in the CF Braun Nitrogen '86 presentation and Fertilizer Focus October 1987, page 48).

All the above designs, although they solve the thermodynamic problem, are very complex and therefore very expensive.

Ammonia synthesis reactors operate in fact at high pressure, generally not below 80 bár, and more often between 130 and 250 bar, and at a high temperature (400 ÷ 500 °C). The connecting tubes of the various items of equipment necessary according to the schemes described above (as schematically shown in the above-mentioned references) operate under critical conditions (high gas temperature between the various reaction beds) and must therefore be made of special material and with long runs to minimize mechanical stress producted by thermal expansion. The situation is particularly complex in reactors according to CF Braun, in spite of the measures taken according to the CF Braun UK application No. 2132501A.

The Applicant, continuing its research in this field, has found a process and reactor with several catalytic beds free from the disadvantages described above, which can be constructed in a single apparatus, permitting the easy exchange of heat between catalytic beds, and in particular before the last catalytic bed, in order to achieve maximum recovery of reaction heat at the highest possible degree of heat, said heat being recovered for example to pre-heat the boiler water of for the direct production of steam.

The main features of the invention are described in the claims.

In an advantageous embodiment, the hot gas reacted in the last catalytic bed but one is transferred, through a duct usually placed axially in a vertical reactor, directly to the heat recovery system (pre-heater or boiler), to be then returned directly to the reactor by means of a duct either external or internal to the above-mentioned transfer duct, creating an airspace for the gas returning to the reactor, which gas then feeds directly the last catalytic bed with an axial-radial or radial flow either centripetal or centrifugal. Said gas, after being reacted in the last catalytic bed, is then transferred again to the central or external part of the reactor, leaving then from the bottom of the reactor.

For a clear understanding of the invention, it is now described with reference to Figure 1, which shows a cross-section of the converter according to a preferred embodiment of the invention.

The reactor consisting of shell (1) and of cartridge (2) forming the catalytic beds (in this case three, 6, 7, 13) is fed the fresh gas (3) entering from the bottom of the reactor and flowing from the bottom towards the top along the airspace (4) between the internal wall of the shell (1) and the cartridge (2) external wall, for the purpose of reducing to a minimum the temperature of the shell.

The gas (3′) leaving said airspace at the upper end of the cartridge containing said three catalytic beds enters the tubes of the gas/gas exchanger (5) situated in the central part of the first catalytic bed (6), where it is pre-heated with the heat from the hot gas (3″) coming from the first catalytic bed (6), said bed being run through by the gas with a centripetal flow (Arrows F1).

The hot gas, after having run along the exchanger (5) on the outside of the tubes, feeds the second catalytic bed (7) running through the bed still with an inward flow (Arrows F2).

The operation of a catalytic bed with indirect exchanger (5) is described in greater detail in the Applicant's US Patent No. 4,405,562.

The hot gas reacted in the second catalytic bed (7) is collected in 15 and is transferred through a central duct (8) situated axially to the reactor into 8′, directly to the heat transfer system (SAC) which is preferably a tube nest (9) with U-shaped tubes (10) where part of the reaction heat is removed by generating steam (11). The gas (20′), after the heat has been removed, returns directly to the reactor through a duct (12) outside the said transfer duct (8), said gas (20′) feeding directly the last catalytic bed (13) running through it with an outward flow (Arrows F3).

The hot reacted gas leaving the bed (13) is then collected in the peripheral external area (30) of said bed and is again transferred to the central part of the reactor running through the airspace (14) of the double bottom of the last catalytic bed, leaving then from the bottom of the reactor through duct (15) (Arrows U).

In the embodiment shown in Figure 2, the gas runs through the catalytic beds with an axial-radial flow, as described in previous Patents granted to the Applicant, such as for example in Swiss Patent No. 643752.

The system for the recovery of heat according to this invention can also be adopted in reactors with catalytic beds in which the gas flow is entirely radial.

The system according to this invention can also be used in reactors operating at a lower pressure where there is no airspace (4) as shown in Figure 1, and the catalytic beds are directly in contact with the internal wall of the reactor shell (1). In this case the hot reacted gas leaving the bed (13) is then sent again to the central part of the reactor through the airspace (14) formed between the bottom of the last catalytic bed and the shell.

The system according to this invention can also be used to modernize reactors for example according to the Applicant's European Patent Publication No. 0202454.

According to the above embodiment (Figure 2), the gas coming from the third bed (7) is now sent to heat exchange system SAC (external exchanger) through an airspace (16) formed between the cartridge wall (2) and a new additional cylindrical wall (17) introduced next to the gas distribution wall (18) feeding the last catalytic bed (13) with a centripetal flow.

After running through the airspace (16), the gas runs through the airspace (19) and enters (Arrows F6) the U-shaped tubes (10) of the exchanger (9).

When leaving the exchanger (10) (Arrow F7) the gas enters the reactor again (Arrow 8) through the duct (10) forming the airspace (19) to feed the catalytic bed (13) through the airspace (21) formed by the closed bottom (35) of the catalytic bed (22) and by the bottom (23) connected with the wall (17).

Wall (17) in its upper part also has lid (24) which prevents the mixing of the gas coming from the catalytic bed (7) with the gas feeding catalytic bed (13).

The gas from catalytic bed (13) runs through the transfer duct (8) and leaves the reactor substantially as in Figure 1.

Claims

1. Process for exothermic heterogeneous synthesis in which the synthesis gas flows through a series of catalytic beds stacked but separate one from the other contained within the same reaction space where the gas leaving one bed runs through the next catalytic bed, characterized by the fact that the hot gas leaving the last catalytic bed but one is transferred outside the space containing the series of beds, where its heat is recovered, and is then re-introduced inside the reaction space, is reacted in the last catalytic bed, and then leaves said reaction space.

2. Process according to claim 1, characterized by the fact that the reaction gas runs with an axial-radial or radial flow, from the outside to the inside, through the catalytic beds from the first to the last but one, and with an outward flow through the last bed.

3. Reactor for the process according to the preceding claims, consisting of an outer pressure-resisting shell, of several baskets for catalytic beds all inside the same shell; of a possible cartridge and of a regenerator, characterized by:
- means for collecting and transferring the hot gas reacted in the last catalytic bed but one after being reacted in the beds going from the first to said last but one, with a centripetal radial flow, to a regenerator situated outside said shell;
- means for returning said gas, after the heat has been recovered in the external regenerator, inside the shell, and to react it with a centrifugal radial flow in the last catalytic bed;
- and means for expelling said gas collected at the perifery of said last catalytic bed from the bottom of the reactor.

4. Reactor according to claim 3, in which the hot gas from the last catalytic bed but one is collected in a first central duct axial to the reactor and taken directly inside the tubes of the regenerator arranged below and at right angles with the reactor, and the gas leaving said tubes is taken to the last bed by a second duct coaxial and external to said first duct.

5. Reactor according to the preceding claim, characterized by an airspace in the double bottom of the last bed to collect and expel centrally from the bottom of the reactor the gas collected at the perifery of the last catalytic bed.

6. Reactor according to the preceding claims, obtained by converting axial reactors into substantially radial reactors, characterized by the fact that the gas from the last bed is taken to external exchanger through an airspace obtained between the wall of the cartridge and an additional wall introduced next to the gas distribution wall in the last catalytic bed run through centripetally.

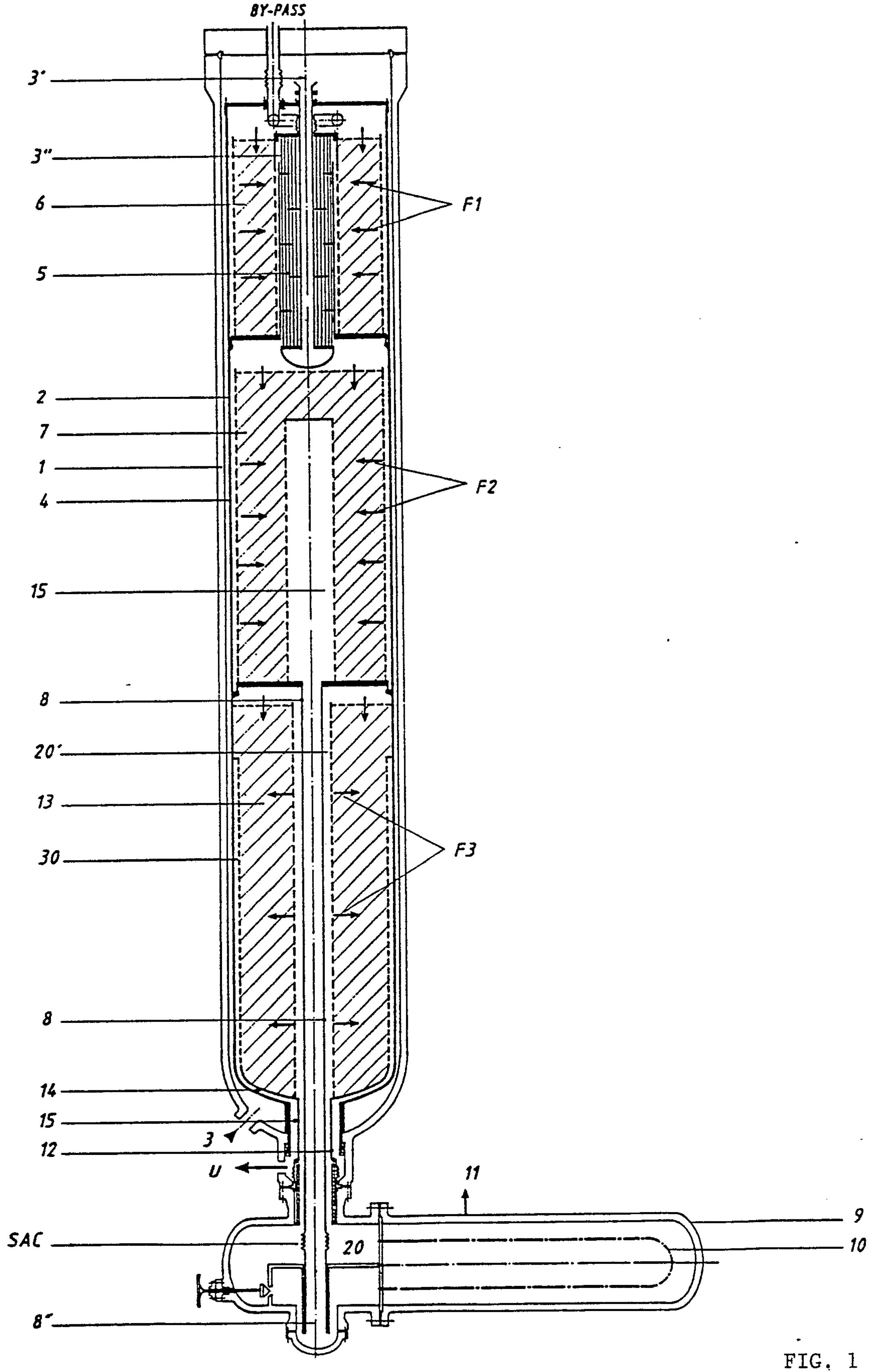
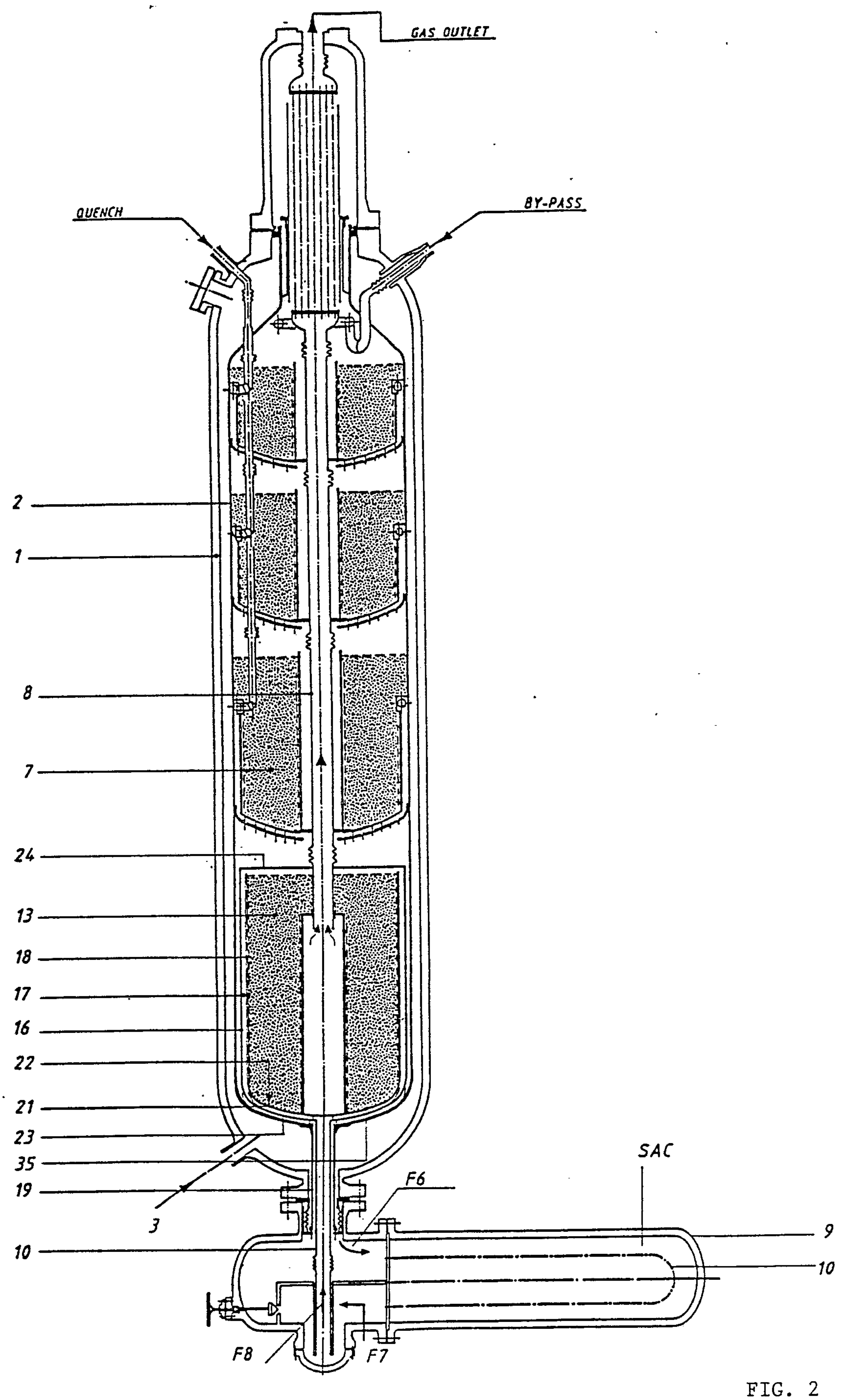


FIG. 1


GAS OUTLET
QUENCH
BY-PASS
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F6
SAC
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F8
F7

FIG. 2

European Patent Office

EUROPEAN SEARCH REPORT

Application Number

EP 89 11 1129

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
A	FR-A-2 183 985 (INSTYTUT NAWOZOW SZTUCZNYCH) * Page 1, lines 1-3; page 2, line 35 - page 4, line 37; figures 1,2 * ---	1,3,6	B 01 J 8/04 // C 01 C 1/04
A	EP-A-0 269 854 (AMMONIA CASALE) * Abstract; claims 1,2,6; figure 2 * -----	1,3	

TECHNICAL FIELDS SEARCHED (Int. Cl.5)
B 01 J C 01 C

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	28-11-1989	SIEM T.D.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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D : document cited in the application
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