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(54) A CATALYTIC REACTOR AND A METHOD OF PREVENTING FLUIDIZATION OF A CATALYST BED IN SUCH A REACTOR.

(71) We, UNITED TECHNOLOGIES CORPORATION, a Corporation organized and existing under the laws of the State of Delaware, United States of America, having a place of business at 1, Financial Plaza, Hartford, Connecticut, 06101, United States of America, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed to be described in and by the following statement:—

This invention relates to a catalytic reactor and to a method of preventing fluidization of a catalyst bed in such a section.

Catalyst reaction apparatus for converting hydrocarbon fuels to useful industrial gases, such as hydrogen, are well known in the art. In one type of apparatus, such as apparatus for steam reforming a hydrocarbon fuel, the process fuel is passed through a tube containing the catalyst. If the reaction is endothermic the tube is usually disposed within a furnace which provides the heat to drive the reaction. If the catalyst filled tube is vertical and the process gas flows upwardly therethrough, the upward force of the flowing gas, particularly at higher through-puts, is usually greater than the weight of the catalyst particles, resulting in continuous motion of the catalyst particles relative to each other. When this condition exists the catalyst bed is said to be fluidized. This continuous motion results in damage to the particles such as causing them to break up into smaller pieces or to wear by the friction of the particles rubbing against each other. Fine size particles are lost by being carried out of the bed with the reacting gases; and, over a period of time, the total volume of catalyst within the bed may be reduced to an unacceptable level, requiring replenishment of the bed.

Anti-fluidizing devices are well known in the art. One common type is a spring loaded perforated member positioned on top of the bed thereby maintaining the bed under compression at all times to prevent fluidization. However, mechanisms of this type are expensive and it is often difficult if not impossible to find a spring material which will withstand the environment of the apparatus in which it is to be used.

Complexity and expense are the major disadvantages of other mechanisms which might be useful in preventing fluidisation.

An object of the invention is to provide an upflow catalytic reactor having relatively uncomplicated and inexpensive means to prevent fluidisation of the catalyst bed.

According to this invention a catalytic reactor comprises: wall means defining a vertically extending reaction chamber having a lower end and an upper end, said lower end including an inlet and said upper end including an outlet, characterised in comprising catalyst particles disposed within said chamber forming a vertically extending catalyst bed therein, said bed having been mechanically presettled; perforated plate means resting by its weight atop said catalyst bed and in contact with the top layer of catalyst particles within said bed, said plate means being of such a weight and having such pressure drop characteristics as to permit expansion of said catalyst bed as a result of upward fluid flow through said bed while remaining in continuous contact with said top layer during expansion and also during contraction of said catalyst bed; and restraint means associated with said reactor and selectively located above said plate means to stop upward movement of said plate means at a predetermined location thereby preventing expansion of said catalyst bed beyond a predetermined volume during operation of said reactor, said predetermined location being selected to stop expansion of said bed prior to the onset of fluidisation.

Also according to this invention a method, for preventing fluidisation of a catalyst bed in a catalytic reactor having a vertical column of particulate catalyst in which gases flow upwards through the column, comprises the steps of:— mechanically presettling the catalyst column prior to operation of the reactor;

placing atop the presettled catalyst column perforated plate means which extends thereacross at a first vertical location and rests thereon by its weight alone, the plate, by its weight and pressure drop characteristics, moving downwardly and upwardly in continous contact with the top layer of catalyst as the catalyst column contracts and expands during operation of the reactor; and limiting upward movement of the plate means to, at most, a point beyond the first vertical location which point is such as to stop expansion of the bed just prior to the onset of fluidisation.

The prior art suggests that in order to prevent fluidisation of a particulate bed, be it a catalyst bed or any other type of bed, the entire bed must be kept under compression. It has been found that this is not so, and that fluidisation does not occur if only the top layer of particles is prevented from lifting off the surface of the bed, as long as only a small expansion of the bed is permitted.

The basic problem with upward flow through beds of particles is the result of a pressure drop through the bed which exceeds the weight of the particles within the bed. At this point the top layer of particles in the bed begins to lift, vibrate, and tumble. As the flow of gas through the bed is increased this particle activity progresses downwardly through the bed until large sections of the bed begin to lift and tumble in a "boiling fashion. This is called fluidization. It has been found that a perforated plate or screen which 1) simply sits on top of the bed in contact with the top layer of catalyst particles, 2) is not heavy enough to prevent expansion of the bed, and 3) is constructed such that the bed expands without the member first lifting off the bed, can prevent fluidization if expansion of the bed is limited to only a few percent of its nonexpanded volume. A simple restraining device or stop is positioned within the reactor whereby the perforated member is limited in its upward movement by coming into contact with the restraint.

The invention will now be described by way of example, with reference to the drawings, in which:—

Figure 1 is a fragmentary, vertical, cross-sectional view of steam reforming reactor apparatus according to the present invention;

Figure 2 is a cross-sectional view of the apparatus of Figure 1 taken substantially along the line 2-2 in Figure 1; and

Figure 3 is an enlarged view of the upper portion of one of the reactors of Figure 1 showing details of the present invention.

Consider, as an exemplary embodiment of the present invention, the catalytic reaction apparatus 10 of Figures 1-3. In this embodiment, the apparatus is for the purpose of steam reforming a reformable hydrocarbon fuel in the presence of a suitable catalyst in order to produce hydrogen. The apparatus 10 comprises a furnace 12 including burner nozzles 14, a burner fuel manifold 16, and an air manifold 18. Disposed within the furnace 12 are a plurality of tubular reactors 20. In this embodiment there are nineteen reactors arranged as shown in Figure 2.

Each reactor 20 comprises an outer cylindrical wall 22 and an inner cylindrical wall or center tube 24 defining an annular reaction chamber 26 therebetween. The reaction chamber 26 is filled with steam reforming catalyst particles or pellets 28 which are supported on a screen 30 disposed at the inlet 32 of the reaction chamber. Any suitable steam reforming catalyst, such as nickel, may be used to fill the reaction chamber. Antifluidization means 33 is disposed at the outlet 36 (Fig 3) of the reaction chamber and is hereinafter more fully explained in conjunction with the more detailed view of Figure 3. The cylinder which is defined by the outer wall 22 is closed at its upper end 38 by an end cap 40. The center tube 24 has an upper inlet end 42 and a lower outlet end 44. The inlet end 42 terminates below the end cap 40 such that the center tube is in gas communication with the outlet 36 of the reaction chamber 26.

Disposed within the center tube is a cylindrical plug 46 which has an outer diameter somewhat smaller than the inner diameter of the center tube thereby defining an annular regeneration chamber 48 therebetween having an inlet 49. The plug 46 may be a solid rod, but in this embodiment is a tube which is blocked by an end cap 50 at one end thereof such that reaction products exiting the reaction chamber 26 must flow around the plug 46 through the regeneration chamber 48. Spacing between the plug 46 and the center tube 24 is maintained by dimples 52 in the plug wall. For the purposes of the present embodiment, the function of the regeneration chamber 48 is to return heat from the reaction products leaving the outlet 36 back into the catalyst bed of the reaction chamber 26. The arrangement shown in Figure 1 provides some preheating of the process fuel before it enters the catalyst bed.

Each reactor 20 may be considered to comprise an upper portion 56 and a lower portion 58. The upper portion 56 is disposed within what is hereinafter referred to as the burner cavity 60. The cavity 60 is that volume of the furnace 12 within which actual combustion of the fuel and air fed into the furnace takes place. This volume is characterized by very high temperatures, considerable radiant heating at well as convective heating of the reactors 20, and axial (i.e., in the direction of the axis of the reactors 20), as well as radial mixing of the gases therein.

The lower portion 58 of each reactor is surrounded by a cylindrical wall or conduit 62 spaced outwardly from the wall 22 defining an annular burner gas passageway 64 therebetween having an inlet 66 and an outlet 67. The outlet 67 is adjacent the inlet 32 of the reaction chamber 26. The passageway 64 is filled with a heat transfer packing material such as spheres 70 of alumina supported on a screen 68. The space 72 between adjacent conduits 62 is filled with a nonheat conductive material such as ceramic fiber insulation which is supported on a plate 74 extending across the furnace and which has holes therein through which the reactors 20 pass. The plate 74 and the material within the space 72 prevents the furnace gases from flowing around the outside of the conduits 62.

In addition to the plate 74, plates 76, 78 and 80 also extend across the furnace and define manifolds therebetween. The plate 80 rests on the bottom wall 82 of the furnace. The plates 78 and 80 define a reaction products manifold 84 therebetween; the plates 76 and 78 define a process fuel inlet manifold 86 therebetween; and, the plates 74 and 76 define a furnace gas outlet manifold 88 therebetween. The plugs 46 and the centre tubes 24 abut the bottom plate 80; the outer walls 22 of the reactors abut the plate 78; and, the conduits 62 abut the plate 74.

In operation, a mixture of steam and reformable hydrocarbon fuel from the manifold 86 enters the inlet 32 of the reaction chamber 26 by way of the holes 100 in the wall 22; the manifold 86 is fed by a conduit 102. The mixture immediately begins to be heated by the furnace gases flowing countercurrent thereto through the passageway 64 and begins to react in the presence of the catalyst particles 28. As the fuel, steam and reaction products travel upward within the reaction chamber 26 they continue to react and pick up additional heat. The hot reaction products enter the inlet 49 of the regeneration chamber 48. As the reaction products traverse the length of the annular regeneration chamber, heat is transferred therefrom back into the reaction chamber 26. They thereupon enter the reaction products manifold 84 through the holes 104 in the center tube 24, and are carried away from the reactor via the conduit 106 either for further processing, storage, or consumption.

Fuel for the furnace enters the manifold 16 via a conduit 108 and thereupon passes into the burner cavity 60 by way of the nozzles 14. Air enters the manifold 18 via a conduit 110 and enters the burner cavity 60 via annular passageway 112 surrounding each nozzle 14. Burning of the fuel and air takes place within the burner cavity 60. The hot gases from the burner cavity travel through the passageways 64 into the manifold 88 and are exhausted via a conduit 113.

Referring to Figure 3, the anti-fluidization means 33 is comprised of a plate 90 and a restraint 92. The plate 90 is an annulus which simply rests by its own weight atop the catalyst bed in contact with the top layer of catalyst pellets. The plate is perforated to permit reaction products to pass therethrough. Its weight and pressure drop characteristics are such that it will not lift off the catalyst bed but will maintain continuous contact with the top layer of catalyst pellets during operation even as the bed expands and contracts. This prevents the top layer of catalyst pellets from becoming fluidized, which in turn prevents the catalyst bed from becoming fluidized, as long as the catalyst bed is not permitted to expand by more than a few percent of its volume (or axial length, which is directly proportional to volume). If the bed is permitted to expand too much, pellets or particles below the top layer will become fluidized, and this fluidization will be transmitted through the bed until the entire catalyst bed is fluidized.

The restraint 92 is provided to limit expansion of the catalyst bed by stopping upward movement of the plate 90. In this embodiment the restraint 92 includes an annular plate 91 welded around its inner circumference 94 to the inside surface of the center tube 24. The plate 91 is perforated with holes 93 therethrough. A cylindrical flange 96 is integral with the plate 91 and extends vertically downwardly therefrom near its outer edge 98. The plate 91 could have been welded to the outer cylindrical wall 22 rather than the center tube 24; however, axial thermal expansion of the center tube 24 is less than that of the outer wall 22 so that it is preferred to fix the plate to the center tube.

As used in this specification and appended claims, the "weight" of the perforated plate includes the weight of anything which may rest on or be attached to or a part of the plate but which moves as one with the plate. For example, although not shown, an annular ring may be secured to the top surface of the plate to provide additional weight. Upward movement of the plate may be stopped when the ring contacts a suitable restraint.

In practicing the present invention it must be kept in mind that, even without fluidization, the catalyst bed will settle during operation of the reactor due to operational vibration as well as thermal expansion and contraction of the outer wall 22 relative to the center tube 24. Therefore, it is desirable to minimize, to the extent possible, settling to the catalyst bed during operation. This is accomplished by presetting the catalyst bed, such as by mechanical vibratory apparatus, as one fills the reaction chamber 26 during assembly of the apparatus.

By presettling the catalyst bed, adding catalyst particles to fill the void left by the presettled particles, presettling the bed again, etc., the reaction chamber can be filled to the extent desired and settling during operation (assuming fluidization is prevented) will thereby be minimized. Once the reaction chamber is filled to the desired level with presettled catalyst particles the perforated plate 90 is placed atop the bed and the restraint 92 is welded into position. Preferably the restraint 92 is located such that the flange 96 is in contact with the plate 90 or as close thereto as possible, recalling that the bed expansion will be the sum of the distance between the plate 90 and the flange 96 plus the amount of contraction of the bed due to unavoidable additional settling during operation of the reactor.

In one test a reactor constructed in all important respects like the reactor shown in Figures 1-3 was operated for a period of 732 hours without any indication that fluidization of the catalyst bed had occurred. In the apparatus tested the annular reaction chamber had an outer diameter of 22.22 cm, an inner diameter of 16.76 cm, and a presettled catalyst bed length of 160.0 cm prior to operation of the reactor. The catalyst was in the form of cylindrical pellets. The plate 90 was made from "Incoloy" (Registered Trade Mark) 800 alloy steel (from International Nickel Co.), was 0.31 cm thick, and had inner and outer diameters of 16.81 cm and 21.71 cm, respectively. It was perforated over its entire surface with 0.31 cm diameter holes to the extent that the plate was 40% porous. The annular plate 91 of the restraint 92 had an outer diameter of 21.71 cm and was 40% porous. The flange 96 was 0.96 cm, long, and prior to operation of the reactor it was essentially in contact with the plate 90. After 732 hours of operation and thirty-four shutdown cycles (i.e. the apparatus is cooled to ambient temperature) settling of the bed amounted to only about 2% of the bed length; no further significant settling is expected to occur with additional operation. Thus, maximum expansion of the bed amounted to about 2% of the bed length and fluidization did not occur. Increase in bed pressure drop was limited to less than 4%. It is believed that if the catalyst bed is presettled, restraining the plate from moving upwardly substantially beyond its initial location will always prevent fluidization since further settling of the bed during operation will always be within acceptable limits.

It is not known just how much expansion can be tolerated without fluidization occuring. From the foregoing example it is apparent that 2% expansion is acceptable. Perhaps up to 5% expansion will be acceptable. If, prior to operation of the reactor, the restraint 92 is located at some distance from the plate 90 (rather than being located as close to the plate 90 as possible) then the amount of expansion during operation will be that much greater. There is no particular advantage in keeping the expansion to an absolute minimum as long as expansion is stopped prior to the onset of fluidization.

It should be apparent that the reactor described in conjunction with the antifluidization means of the present invention is by way of example only. For instance, the invention is as useful for preventing fluidization of a cylindrical catalyst bed as it is for the annular bed hereinabove described. It should also be apparent that the invention is not limited to use with steam reforming catalyst beds.

Features of the apparatus described above are claimed in our co-pending application No. 48121/77, Serial No. 1,545,669.

WHAT WE CLAIM IS:–

1. A catalytic reactor comprising: wall means defining a vertically extending reaction chamber having a lower end and an upper end, said lower end including an inlet and said upper end including an outlet, characterized in comprising catalyst particles disposed within said chamber forming a vertically extending catalyst bed therein, said bed having been mechanically presettled; perforated plate means resting by its weight atop said catalyst bed and in contact with the top layer of catalyst particles within said bed, said plate means being of such a weight and having such pressure drop characteristics as to permit expansion of said catalyst bed as a result of upward flow through said bed while remaining in continuous contact with said top layer during expansion and also during contraction of said catalyst bed, and restraint means associated with said reactor and selectively located above said plate means to stop upward movement of said plate means at a predetermined location thereby preventing expansion of said catalyst bed beyond a predetermined volume during operation of said reactor, said predetermined location being selected to stop expansion of said bed prior to the onset of fluidization.

2. A reactor according to claim 1, characterized in that said restraint means is fixedly attached to said wall means.

3. A reactor according to claim 1, characterized in that said restraint chamber is an annular reaction chamber and said wall means includes an inner cylindrical wall and an outer cylindrical wall spaced apart to define said annular reaction chamber, and said perforated plate means is a first annular plate.

4. A reactor according to claim 3 characterised in that said restraint means is fixed to said inner cylindrical wall.

5. A reactor according to claim 3 or claim 4 characterised in that said restraint means includes a perforated second annular plate having an inner circumference and an outer circumference, said inner circumference being secured to said inner cylindrical wall, said second annular plate including a cylindrical flange extending vertically toward said first annular plate, and said flange being engageable with said first annular plate to limit upward movement thereof.

6. A method for preventing fluidisation of a catalyst bed in a catalytic reactor having a vertical column of particulate catalyst in which gases flow upwardly through the column, the method comprising the steps of:– mechanically presettling the catalyst column prior to operation of the reactor; placing atop the presettled catalyst column perforated plate means which extends thereacross at a first vertical location and rests thereon by its weight alone, the plate,

by its weight and pressure drop characteristics, moving downwardly and upwardly in continuous contact with the top layer of catalyst as the catalyst column contracts and expands during operation of the reactor; and limiting upward movement of the plate means to, at most, a point beyond the first vertical location which point is such as to stop expansion of the bed just prior to the onset of fluidisization.

7. The method according to claim 6 wherein the said step of limiting includes limiting upward movement of said plate means to substantially said first vertical location.

8. A catalytic reactor constructed and arranged substantially as herein described and shown in the drawings.

9. A method for preventing fluidisation of a catalyst bed in a catalytic reactor substantially as herein described with reference to the drawings.

WITHERS & ROGERS
Chartered Patent Agents
4, Dyer's Buildings,
Holborn, London,
EC1N 2JT
Agents for the Applicant

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1564994 COMPLETE SPECIFICATION

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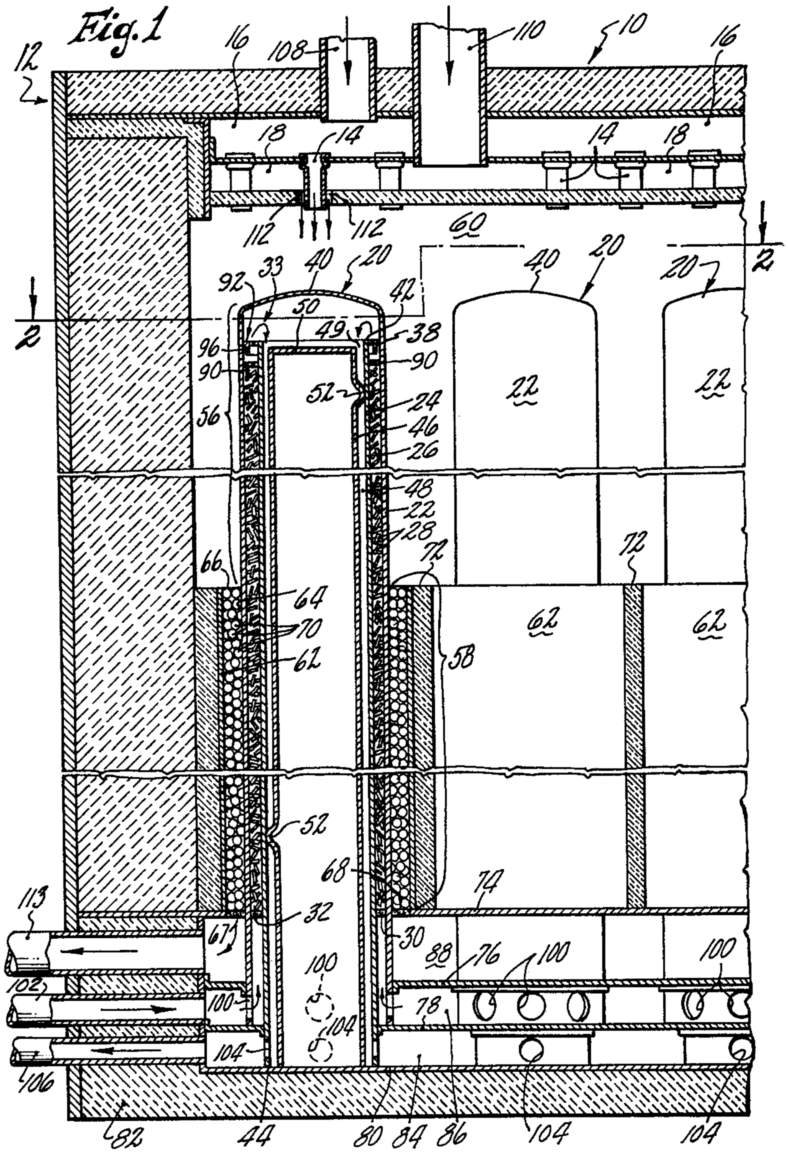
Sheet 1

Fig. 1



1564994 COMPLETE SPECIFICATION

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Sheet 2

Fig. 2

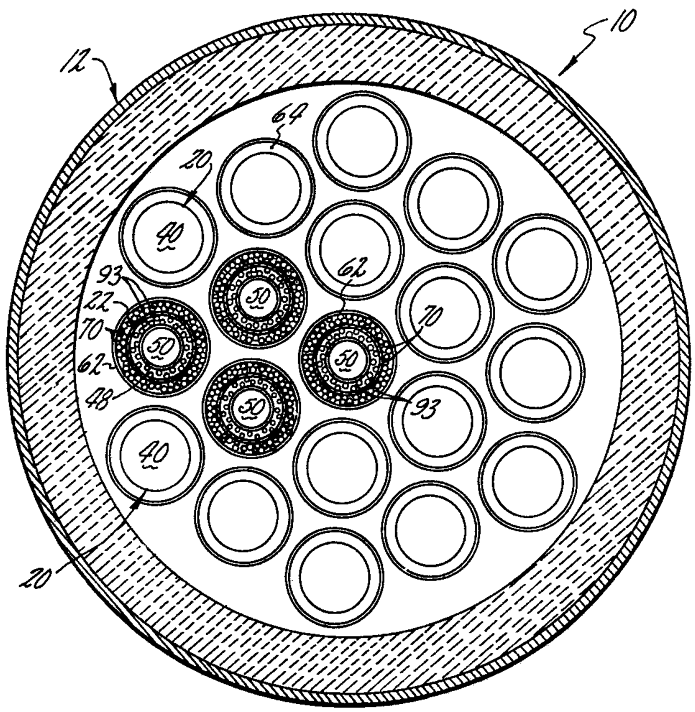
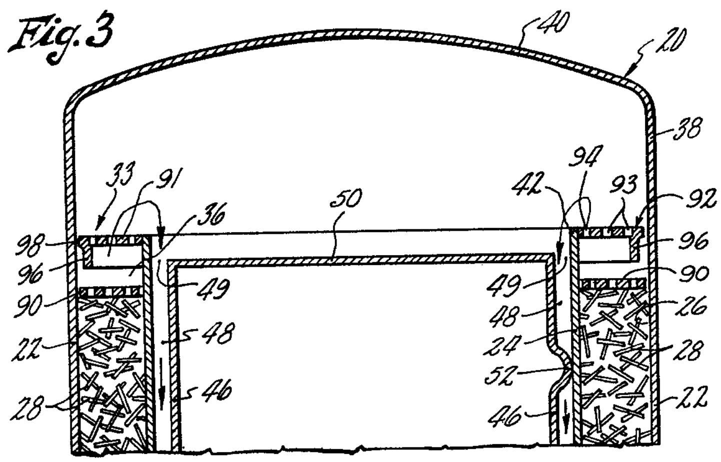


Fig. 3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