

PATENT SPECIFICATION (11)

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(54) A PROCESS FOR FILLING A REACTOR WITH SOLID PARTICLES

(71) We, SHELL INTERNATIONALE RE-
 SEARCH MAATSCHAPPIJ B.V., a company
 organised under the laws of The Nether-
 lands, of 30 Carel van Bylandtlaan, The
 Hague, The Netherlands, 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 par-
 ticularly described in and by the following
 statement:—

The present invention relates to a process
 for filling a reactor with solid particles to
 form one or more homogeneous beds of
 solid particles, in particular catalyst par-
 ticles, therein.

In the technical application of reactions in
 which liquids and/or gases are passed over
 a particulate catalyst it is generally of im-
 portance that the catalyst particles should
 be present in one or more homogeneous
 beds, i.e. beds having a uniform packing and
 an arbitrary orientation of the catalyst par-
 ticles. Consequently, a regular flow through
 the catalyst bed and an optimum degree of
 wetting of catalyst particles by the reaction
 mixture are obtained, and moreover no re-
 action mixture leaves the reactor without
 having contacted the catalyst or after too
 short a contact period.

Although the filling of relatively small re-
 actors with catalyst particles to form therein
 a homogeneous bed of catalyst particles (usu-
 ally consisting of small cylinders) is diffi-
 cult, it can be achieved by pouring the cata-
 lyst particles carefully into the reactor, or
 by introducing the catalyst particles slurried
 in a liquid into the reactor. Much greater
 problems arise when large reactors (for ex-
 ample, consisting of cylinders having a dia-
 meter exceeding 2 m and a height exceeding
 10 m) have to be provided with one or more
 homogeneous beds of catalyst particles.

Large reactors of this type can be used,
 for example, in processes for the hydrode-
 sulphurization of mineral oil fractions, in
 particular of crude mineral oil or residues
 obtained by removing at least part of the
 volatile products from a crude mineral oil.

The term "mineral oil" is understood to in-
 clude petroleum, shale oil, rock oil, oil ob-
 tained from tar sands and similar hydro-
 carbon-based natural oils present in the
 earth's crust.

The present invention provides a process
 by means of which a reactor can be provided
 with one or more homogeneous beds of solid
 particles, for example, catalyst particles.

According to the present invention there
 is provided a process for filling a reactor
 with solid particles to provide one or more
 homogeneous beds of solid particles there-
 in, in which solid particles in the reactor
 are brought into a state of incipient fluidiza-
 tion by means of a liquid stream, after which
 the rate of flow of the liquid stream is re-
 duced or the liquid stream is discontinued
 and the particles are allowed to settle.

By fluidization is meant a situation in
 which solid particles are suspended in a
 liquid. By maintaining a liquid stream the
 solid particles are prevented from settling;
 they are in a fluidized bed. The expansion
 of the fluidized bed means the percentage
 by which the volume of the fluidized bed ex-
 ceeds the volume of the bed formed by the
 fully settled catalyst particles. By the term
 "incipient fluidization" is meant that the de-
 gree of this expansion is low, i.e. below 10%,
 preferably below 5%. It will be clear that
 the lower the degree of expansion of the
 fluidized bed, the more complete will be the
 filling of the reactor after the settling of
 the catalyst particles (and consequently the
 less empty space will be present in the re-
 actor).

The rate of flow of the liquid stream which
 must be applied in order to maintain cata-
 lyst particles of a given type having given
 dimensions in a state of incipient fluidiza-
 tion, must be predetermined in model tests.

It is possible to fill the empty reactor in
 advance with solid particles and subse-
 quently to introduce a liquid stream into the
 reactor in order to bring the particles into
 a state of incipient fluidization. Filling the re-
 actor with solid particles (for example, by

means of a hopper) is a time-consuming operation and moreover a high degree of breakage of the solid particles will occur. Therefore, the solid particles are preferably introduced into the reactor as a slurry in a liquid.

While the solid particles (which may or may not be in the form of a slurry) are being introduced, the liquid stream to bring the particles into a state of incipient fluidization may or may not be maintained. If the said liquid stream is maintained during the filling with the solid particles (which is preferred), the latter are very suitably supplied at the top of the reactor.

The reactor preferably contains one or more conical grates for the solid particles, so that a bed of solid particles can be formed on each grate. These grates have a central opening through which the solid particles can descend when the reactor has to be emptied, and the inclination of the grates is preferably such that the solid particles can leave the reactor under the influence of gravity when the reactor has to be emptied. A reactor of this type is described in, for example, Netherlands Patent Application No. 7,401,733.

According to the invention, during the filling of the reactor the solid particles are brought into a state of incipient fluidization by means of a liquid stream. In order also to fill the spaces below the grates as well as possible, it is advantageous to decrease the rate of flow of the liquid stream once or several times for a short period shortly before reducing this rate of flow or discontinuing the liquid stream.

The liquid stream used is preferably a stream of a petroleum-based product. Petroleum distillate fractions, in particular gas oil, are very suitable.

Although it is possible to supply fresh liquid continuously in order to bring the solid particles into a state of incipient fluidization, the liquid will generally be recycled. The liquid stream is very suitably introduced at the bottom of the reactor at the desired rate and is discharged at the top of the reactor, preferably at a place where solid particles are no longer present in the liquid.

When the reactor contains all the solid particles to be introduced in a state of incipient fluidization, the rate of flow of the liquid stream is reduced or the stream is discontinued and the particles are allowed to settle. After settling the liquid can optionally be discharged or replaced by an other liquid. In certain cases the same liquid used for the fluidization can be passed at elevated temperature and/or pressure over the homogeneous catalyst bed now obtained in order to give the catalyst an active form (for example in sulphided form for desulphurization catalysts based on supported

metals of Group VI and/or Group VIII).

In order to obtain a solid-particle bed which does not show a very high pressure drop when a liquid to be converted by these particles is passed over the bed, it is advantageous that the dimensions of the solid particles remain within narrow limits. Especially the presence of solid particles having smaller dimensions than desired and of fines should be avoided, because they can clog the passages and consequently cause a substantial pressure drop.

In the production of catalyst particles, for example by extrusion, breakage usually occurs during their further treatment (transport, pouring etc.), which results in the formation of smaller particles, while a quantity of pulverulent material (fines) is always present.

The solid particles the dimensions of which lie within narrow limits and which, as described above, are very suitable to be incorporated in the solid-particle bed in the reactor, are preferably obtained by fluidizing a mixture comprising, in particular consisting of, particles the dimensions of which lie within narrow limits and of particles having smaller dimensions in a liquid until the particles of smaller dimensions have accumulated in the upper layer, then discontinuing the fluidization and removing the upper layer of settled particles, which layer consists of particles having smaller dimensions. These smaller particles are very suitably removed by suction. During the said fluidization, which is very suitably performed in a supply tank (a large, for example cylindrical vessel) the fines usually do not settle, but are entrained by the liquid and removed from the supply tank. Before supplying the fluidization liquid again at the bottom of the supply tank, the fines are very suitably removed by filtration.

The use of the said supply tanks moreover has the great advantage that they can be used as storage space for the solid particles. After the solid particles the dimensions of which lie within narrow limits have been obtained in the supply tank, they can be supplied to the reactor at any desired moment, preferably as a slurry as described before.

When the solid particles in the reactor have to be replaced, this operation can be carried out very rapidly. The reactor is emptied and then according to the invention filled from the supply tank with particles the dimensions of which lie within narrow limits, which operation can be carried out in a few hours. When the reactor has to be filled with solid particles which have not been collected in advance in a supply tank or otherwise, this operation may take many days in the case of a reactor of large dimensions. It will be clear that it is favourable

that the supply tank should have a volume of the same order of size (preferably somewhat larger) as that of the reactor.

The solid particles are preferably extrudates. Very suitably they have a diameter of 0.5—3 mm and a length of 3—7 mm.

The solid particles are very suitably catalyst particles, in particular catalyst particles which can be used for the demetallization and/or hydrodesulphurization of crude mineral oil or residues. They preferably contain one or more metals of Group(s) VI and/or VIII of the periodic system or compounds thereof on an oxidic carrier; in particular catalyst particles consisting of one or more oxides and/or sulphides of cobalt, nickel molybdenum and tungsten on a carrier consisting completely or predominantly of alumina and/or silica are very suitable.

The demetallization and/or hydrodesulphurization of for example a crude mineral oil or a residue can be carried out in the reactor under conditions known per se, such as temperatures of 385—445°C. hydrogen pressures of 75—225 kg/cm² and space velocities of 0.5—5 kilograms of feed per cubic metre of catalyst per hour.

The invention will now be described with reference to the accompanying drawing, which is a diagrammatic representation of a suitable apparatus, but is by no means limitative with respect to the scope of the invention.

In the drawing the reference numeral 1 designates a reactor which can be filled with catalyst particles from a supply tank 2. The particles can leave the supply tank 2 through a line 3 and mixed with liquid (for example gas oil) supplied through a line 4, they can be supplied at the top of the reactor 1 through a line 6 by means of a slurry pump 5. At the same time liquid can be supplied at the bottom of the reactor 1 through a line 7 at such a rate that the catalyst particles are brought into a state of incipient fluidization. Line 8 serves to discharge liquid at the top of the reactor. Through this line 8 liquid can be supplied to a reservoir 9 which is provided with a vent line 10. A pump 11 serves to pump liquid from the reservoir 9 to lines 4 and 7 through a line 12. The supply tank 2 comprises a solid-particle inlet 17, a liquid inlet (which is closed when the reactor 1 is filled) connected to line 13 through which, with the lines 3, 4 and 7 closed, liquid can be pumped from the reservoir 9 to the supply tank 2 through line 12 by means of pump 11. The top of the supply tank 2 is provided with a discharge line 14 through which liquid can be recycled to the reservoir 9. If desired, fines can be removed from the liquid in line 14 by means of filter device 15. A line 16 is present at the bottom of the supply tank to remove liquid completely or substantially

completely from the supply tank, which can be performed when the tank has been filled with solid particles the dimensions of which lie within narrow limits.

EXAMPLE

The numbers in this example refer to the drawing. A reactor 1 (height 23 m, diameter 3.4 m) is filled with 170 m³ of extruded catalyst particles having an average diameter of 1.5 mm and an average length of 5 mm. The catalyst particles are supplied from the supply tank 2 (height 26 m, diameter 3.4 m) through a pipe 3, pump 5 and line 6, at a rate of 15 tons/h by means of gas oil which is supplied through the line 4 at a rate of 223 m³/h. Through the line 7 gas oil is supplied to the reactor 1 at a rate in the reactor of 7 mm/sec., with the result that the particles in the reactor are brought into a state of incipient fluidization. From the top of the reactor gas oil is recycled to reservoir 9 through the line 8. When the required quantity of catalyst particles is in the reactor in a state of incipient fluidization, the rate of flow of the gas oil supplied through 7 is briefly decreased a number of times. The supply of gas oil is then discontinued and, if desired, the gas oil present in the reactor can be discharged.

The particles to be transported to the reactor from the supply tank 2 are obtained as follows:

The supply tank is filled with catalyst particles, for example from drums, through the inlet 17. Fines and smaller particles are removed therefrom by fluidizing the contents of the supply tank by means of gas oil which is supplied via the line 13 at a rate in the supply tank of 9 mm/sec. Gas oil is discharged through the line 14 at the top of the supply tank and passed to the reservoir 9 after filtration in the filter device 15. The supply of gas oil is terminated after about 3 hours and the gas oil is discharged from the supply tank. Particles of smaller dimensions, which have collected in the upper layer of the contents of the supply tank, are subsequently removed by means of a large vacuum cleaner.

WHAT WE CLAIM IS:—

1. A process for filling a reactor with solid particles to provide one or more homogeneous beds of solid particles therein, wherein solid particles in the reactor are brought into a state of incipient fluidization by means of a liquid stream, after which the rate of flow of the liquid stream is reduced or the liquid stream is discontinued, and the particles are allowed to settle.
2. A process as claimed in claim 1, wherein the solid particles are introduced into the reactor as a slurry in a liquid.
3. A process as claimed in either of the



that the supply tank should have a volume of the same order of size (preferably somewhat larger) as that of the reactor.

5 The solid particles are preferably extrudates. Very suitably they have a diameter of 0.5—3 mm and a length of 3—7 mm.

10 The solid particles are very suitably catalyst particles, in particular catalyst particles which can be used for the demetallization and/or hydrosulphurization of crude mineral oil or residues. They preferably contain one or more metals of Group(s) VI and/or VIII of the periodic system or compounds thereof on an oxidic carrier; in particular 15 catalyst particles consisting of one or more oxides and/or sulphides of cobalt, nickel molybdenum and tungsten on a carrier consisting completely or predominantly of alumina and/or silica are very suitable.

20 The demetallization and/or hydrosulphurization of for example a crude mineral oil or a residue can be carried out in the reactor under conditions known per se, such as temperatures of 385—445°C. hydrogen 25 pressures of 75—225 kg/cm² and space velocities of 0.5—5 kilograms of feed per cubic metre of catalyst per hour.

30 The invention will now be described with reference to the accompanying drawing, which is a diagrammatic representation of a suitable apparatus, but is by no means limitative with respect to the scope of the invention.

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completely from the supply tank, which can be performed when the tank has been filled with solid particles the dimensions of which lie within narrow limits.

EXAMPLE

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The particles to be transported to the reactor from the supply tank 2 are obtained as follows:

100 The supply tank is filled with catalyst particles, for example from drums, through the inlet 17. Fines and smaller particles are removed therefrom by fluidizing the contents of the supply tank by means of gas oil which is supplied via the line 13 at a rate in the supply tank of 9 mm/sec. Gas oil is discharged through the line 14 at the top of the supply tank and passed to the reservoir 9 after filtration in the filter device 15. The supply of gas oil is terminated after about 3 hours and the gas oil is discharged from the supply tank. Particles of smaller dimensions, which have collected in the upper layer of the contents of the supply tank, are subsequently removed by means of a large vacuum cleaner. 115

WHAT WE CLAIM IS:—

1. A process for filling a reactor with solid particles to provide one or more homogeneous beds of solid particles therein, wherein solid particles in the reactor are brought into a state of incipient fluidization by means of a liquid stream, after which the rate of flow of the liquid stream is reduced or the liquid stream is discontinued, and the particles are allowed to settle. 125

2. A process as claimed in claim 1, wherein the solid particles are introduced into the reactor as a slurry in a liquid.

3. A process as claimed in either of the 130

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- preceding claims, wherein the liquid stream to bring the solid particles into a state of the incipient fluidization is maintained, while the reactor is being filled with the solid particles.
4. A process as claimed in any one of the preceding claims, wherein the solid particles are introduced at the top of the reactor.
5. A process as claimed in any one of the preceding claims, wherein the reactor contains one or more conical grates for the solid particles.
6. A process as claimed in any one of the preceding claims, wherein shortly before reducing the rate of flow or discontinuing the liquid stream, the rate of flow thereof is once or several times decreased for a short period.
7. A process as claimed in any one of the preceding claims, wherein the liquid stream is a stream of a petroleum-based product.
8. A process as claimed in claim 7, wherein the liquid stream is a gas oil stream.
9. A process as claimed in any one of the preceding claims, wherein the dimensions of the solid particles lie within narrow limits.
10. A process as claimed in claim 9, wherein the solid particles the dimensions of which lie within narrow limits are obtained by fluidizing a mixture consisting of particles the dimensions of which lie within narrow limits and of particles having smaller dimensions in a liquid until the particles of smaller dimensions have accumulated in the upper layer, then stopping the fluidization and removing the upper layer of the settled particles, which layer consists of particles of smaller dimensions.
11. A process as claimed in claim 10, wherein the upper layer consisting of particles of smaller dimensions is removed by suction.
12. A process as claimed in any one or more of the preceding claims, wherein the solid particles are extrudates having a diameter of 0.5—3 mm and a length of 3—7 mm.
13. A process as claimed in any one of the preceding claims, wherein the solid particles are catalyst particles.
14. A process as claimed in any one of the preceding claims, wherein the solid particles are catalyst particles which comprise one or more metals of Group(s) VI and/or VII of the periodic system or compounds thereof on an oxidic carrier.
15. A process as claimed in claim 14, wherein the catalyst particles contain oxides and/or sulphides of cobalt and/or molybdenum on a carrier consisting completely or predominantly of alumina.
16. A process as claimed in claim 1 and substantially as described hereinbefore with reference to the Example.
17. An apparatus suitable for carrying out the process claimed in claim 1 and substantially as hereinbefore described with reference to the accompanying drawing.
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