(12) STANDARD PATENT (11) Application No. AU 2008260467 B2
(19) AUSTRALIAN PATENT OFFICE

(54) Title
Apparatus and method for top removal of granular and fine material from a fluidized bed deposition reactor

(51) International Patent Classification(s)
B01J 8/18 (2006.01)

(21) Application No: **2008260467** (22) Date of Filing: **2008.07.10**

(87) WIPO No: **WO08/150552**

(30) Priority Data

(31) Number	(32) Date	(33) Country
111810422	**2007.06.04**	**US**

(43) Publication Date: **2008.12.11**
(44) Accepted Journal Date: **2012.07.19**

(71) Applicant(s)
Lord Ltd. LP

(72) Inventor(s)
Lord, Stephen M.

(74) Agent / Attorney
Davies Collison Cave, 1 Nicholson Street, Melbourne, VIC, 3000

(56) Related Art
US 2007/0040056
US 5798137
US 5876793
US 2004/0151652

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
11 December 2008 (11.12.2008)

PCT

(10) International Publication Number
WO 2008/150552 A3

(51) International Patent Classification:
B01J 8/18 (2006.01)

(21) International Application Number:
PCT/US2008/008569

(22) International Filing Date: 10 July 2008 (10.07.2008)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
111810422 4 June 2007 (04.06.2007) US

(71) Applicant and
(72) Inventor: LORD, Stephen, M. [US/US]; 109 Peppertree Lane, Encinitas, CA 92024-6400 (US).

(81) Designated States *(unless otherwise indicated, for every kind of national protection available)*: AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BR, BW, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IS, JP, KE, KG, KM, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PG, PH, PL, PT, RO, RS, RU, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States *(unless otherwise indicated, for every kind of regional protection available)*: ARIPO (BW, GH, GM, KE, LS, MW, MZ, NA, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MT, NL, NO, PL, PT, RO, SE, SI, SK, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Published:
— *with international search report*
— *with information concerning request for restoration of the right of priority in respect of one or more priority claims; the decision of the receiving Office regarding the request for restoration is pending and will be published separately once available*

(88) Date of publication of the international search report:
12 March 2009

(54) Title: APPARATUS AND METHOD FOR TOP REMOVAL OF GRANULAR FROM A FLUIDIZED BED DEPOSITION REACTOR

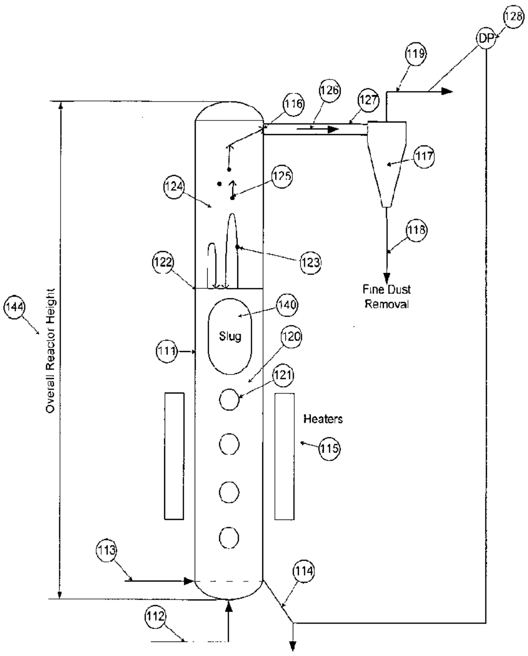

Fig 1. Typical Fluidized Bed Deposition Reactor

(57) Abstract: Apparatus and method for top removal of granular material from a fluidized bed deposition reactor. Removal of the product from the top of the reactor enables a decreased disengaging height and provides a passive means of controlling the bed level despite deposition increasing the weight and height of the bed. The savings from reducing the disengaging height allow use of a taller fluidized bed in a shorter overall reactor length and thus provides increased production with reduced reactor cost. The separation of the gas inlet from the product outlet allows the gas inlet area to be cooler than the product outlet. The separation of the product grinding, caused by the inlet gas, from the product outlet reduces the loss of seed in the product and produces a more uniform product. Removing the hot product and the hot gas at the same place allows energy recovery from both in a single step.

WO 2008/150552 A3

2008260467 04 Jul 2012

Apparatus and method for top removal of granular material from a fluidized bed deposition reactor

Technical Field

This invention relates generally to the field of deposition reactors and more specifically to an apparatus and method for top removal of granular material from a fluidized bed deposition reactor.

Background

Fluidized bed reactors have a long tradition in the chemical industry where the bed usually consists of a finely divided valuable catalyst which makes it necessary to design the reactors to prevent catalyst losses. Thus was developed the practice of requiring a large disengaging height above the bed surface and of using cyclones to capture the fine dust and return it to the bed. A concept called total disengaging height, or TDH, was developed to estimate the height where all the particles that would settle out by gravity had settled out. Internal cyclones were provided at this height to capture the finer dust and return it to the bed. Whenever the catalyst was removed it was removed from the bottom by gravity. Other reactors called dilute phase or transport reactors entrained all the solids up through the reactor and out the top, but these reactors did not have a recognizable bed. When these gas-solids reactor concepts were applied to the design of deposition reactors where gases are introduced to make the bed particles grow, the dilute phase reactor had the major problem that it produced mainly a fine dust which was undesirable. Thus the majority of deposition reactors have been fluidized beds and so the basic design of fluidized beds with a large disengaging height and bottom solids outlet has been used. The idea of internal cyclones has seldom been used because of the deposition on the outside of the cyclones and the problems of reintroducing particles without plugging the cyclone outlets. Since some fine dust is always made, most deposition reactors have external cyclones or filters to trap the dust and prevent damage to the equipment used to recover the effluent gases. Thus the historic approach has been to have removal of the product from the bottom, provide a large disengaging height to minimize product loss, and use external dust removal.

The primary use for deposition reactors is in high purity silicon deposition and Lord in US 6,451,277 in Fig. 1b describes a bed heating method which removes beads from near the top of the bed and then heats them and returns them to the bed. It is notable that the product, **3**, is still removed from the bottom. In the above patent this bed heating method is rejected in favor of a preferred option where the beads are removed by gravity from the bottom then reheated and returned to the bed in a pulsed mode. Lord in

US 6,827,786 provides a detailed description of a multistage deposition reactor which takes advantage of increased bed height to produce additional silicon by use of additional gas injection points along the side of the reactor. In this design the seed generation by grinding is spread out along the reactor because of the extra nozzles and some deposition occurs further from the inlet, but most of the grinding and deposition occurs in the bottom where the solid product is removed. Lord discusses, Col3 line 25, the "De Beers" paper which showed the need for some residence time and temperature to fully crystallize the product and dehydrogenate the beads. He does this in the pulsed bead heater at high temperature and with short residence time. Lord and his many references do not discuss energy recovery from the effluent gas although Lord in US 5,798,137 and 6,451,277 discusses the use of the heat from the outgoing product to heat the incoming gas.

The primary deficiency of the prior technology is staying with the inherited fluid bed design of a bottom outlet and large disengaging space and accepting the inherent conflicting demands caused by introducing the cold deposition gas, which also provides the bulk of the seed generation by grinding, at the same location as the hot product was removed. Lord in various patents attempts to deal with the heat and seed generation problem by spreading out the gas injection, but sufficient gas to fully fluidize the bed must be injected at the bottom, so there is a limit to what can be accomplished in this manner. Inevitably the bottom temperature must be maintained above 800° C to provide the needed crystallization, and some seeds are lost to the product which is in turn contaminated with broken "seed beads." The combination of high temperature and

2008260467 04 Jul 2012

high deposition gas concentration leads to rapid reactions, increased wall deposits and increased risk of agglomeration and plugging.

This multistage design approach also leads to tall reactors and there are cost and manufacturability issues in producing the high purity liners for such reactors which restrict the number of stages and hence production capacity of a given diameter reactor. It is also necessary to measure the bed level and take corrective action by removing some of the bed as the bed grows by opening valves and changing purge flows to allow the right amount of beads to leave the bed. Errors or stuck valves can lead to situations where the bed is too high or too low. Both of these conditions are undesirable upsets.

It is desired, therefore, to provide a method of operation of a fluidized bed deposition reactor for top removal of granular material that alleviates one or more of the above difficulties, or that at least provides a useful alternative.

Summary

In accordance with the present invention, there is provided a method of operation of a fluidized bed deposition reactor for top removal of granular material comprising:

providing a vertical generally cylindrical reactor of a predetermined height, said reactor having at least one gas inlet at or near the bottom thereof and at least one gas and solids outlet at or near the top thereof,

providing at least one gas and granular product separator to separate gas from granular product and a conduit between the gas and solids outlet and said at least one separator,

establishing a heated reaction zone;

providing a fluidizing gas to the reaction zone at a predetermined flow rate,

providing granular particles to the reaction zone to establish the desired fluidized bed, said particles having a variable fluidized height, the bed being fluidized to establish a bubbling fluidized bed with a top of a defined stable height, a disengaging space being provided above said top, the height of said disengaging space being no greater than the distance between the defined top of the bubbling bed and the gas and solids outlet of the container,

providing a second gas to the reaction zone while adjusting the flow rate of the fluidizing gas so that a majority of the granular particles are retained in the reactor while maintaining bubbling, the second gas depositing a coating on said granular particles, thus increasing the size and weight of the particles resulting in an increase in the height of the bed, and

allowing the height of the bed to increase and the disengaging height to decrease until the bursting of bubbles near the surface of the bed periodically throws granular particles out of the gas and solids outlet of the reactor, along the conduit and into the at least one gas and granular product separator,

wherein the flow rates of the fluidizing gas and the second gas are controlled so as to establishing a stable height of the bed.

Embodiments of the present invention provide technical benefits such as passive level control, decreased disengaging height, taller fluidized bed in a shorter reactor, separation of gas inlet from product outlet, separation of product grinding from product outlet and energy recovery which in turn lead to lower capital and operating cost, a better quality product and greater throughput for a given reactor diameter.

2008260467 04 Jul 2012

Brief Description of the Drawings

The drawings constitute a part of this specification and include exemplary embodiments of the invention, which may be embodied in various forms. It is to be understood that in some instances various aspects of the invention may be shown exaggerated or enlarged to facilitate an understanding of the invention.

Figure 1 is a schematic diagram illustrating the operation of a fluidized bed deposition reactor of the prior art with bottom removal and a large disengaging space.

Figure 2a is the same diagram modified to show the benefits of the described embodiments of the present invention.

Figure 2b is a detailed schematic of the top of a reactor showing the granular particle removal mechanism in accordance with an embodiment of the present invention.

Figure 3 is a schematic of a product separator with integrated heat recovery in accordance with an embodiment of the present invention.

Detailed Description

Detailed descriptions of some embodiments of the present invention are provided herein. It is to be understood, however, that the present invention may be embodied in various forms. Therefore, specific details disclosed herein are not to be interpreted as limiting, but rather as a basis for the claims and as a representative basis for teaching one skilled in the art to employ the present invention in virtually any appropriately detailed system, structure or manner.

Turning first to Fig 1 there is shown a schematic of a typical fluidized bed deposition reactor comprising a containment vessel or liner, **111**, of a height, **144**, a gas introduction means, **112**, an optional gas distribution means, **113**, a bottom product removal means, **114**, a bed heating means, **115**, a gas/dust mixture exit, **116**, a connecting means, **127**, a dust/gas separation means, **117**, a dust removal means, **118**, and a gas exit, **119**. The containment vessel, **111**, surrounds a bed of granules, **120**, fluidized by gas bubbles, **121**, and having an average top level, **122**, above which product granules, **123**, thrown up above the bed describe arcs as they rise from random impact within the bed then fall under gravity in a reduced disengaging space, **124**, while the small entrained dust particles, **125**, continue up and leave with the effluent gas, **126**, through the gas/dust mixture exit, **116**, through the connecting means, **127**, then enter the dust/gas separation means, **117**, where most of the dust, **125**, is removed from the gas, **126**, and then ultimately leaves the system via the dust removal means, **118**, while the gas, **126**, and residual dust leaves via an exit, **119**. The differential pressure meter,

128, measures the difference in pressure between the bottom product removal means, **114**, and the gas exit, **119**. This measurement indicates the level, **122**, of the bed of granules, **120**. The bottom removal means, **114**, is used to control the top level, **122**, to maintain the disengaging space, **124**, so that the product granules, **123**, are returned to the bed of granules, **120**, and are thus removed by the bottom product removal means, **114**. This is a very general schematic and the patent literature is full of the various methods and machines that have been proposed to fulfill these requirements. It is possible to have more than one gas entry and to avoid the gas distribution mechanism; the heating means can be of many different kinds, and the dust removal can be done by a cyclone as shown, by a filter or by another gas cleaning device.

In accordance with the present invention, Fig 2a shows a schematic similar to Fig 1 but modified to remove the granular product from the top via a gas/granular separator means, **230**, inserted before the effluent gas enters the gas/dust separation means, **217**. A further modification is the removal of the differential pressure transmitter, **128**, shown in Fig 1, which is not required for bed level control. The invention thus comprises a containment vessel or liner, **211**, of a height, **244**, a gas introduction means, **212**, an optional gas distribution means, **213**, an optional bottom product removal means, **214**, a bed heating means, **215**, a gas/dust/granular mixture exit, **216**, a first connecting means, **241**, a gas/granular separator means, **230**, with a granular removal means, **231**, an optional heat recovery means, **242**, a further connecting means, **229**, a gas/dust separation means, **217**, a further optional heat recovery means, **243**, a dust removal means, **218**, and a gas exit, **219**. The containment vessel, **211**, surrounds a bed of granules, **220**, fluidized by gas bubbles, **221**, and slugs, **240**, and having an average top level, **222**, above which some granules, **223**, thrown up above the bed describe arcs as they rise from random impact within the bed then fall under gravity in a reduced disengaging space, **224**, while some granules, **236**, and the small entrained dust particles, **225**, continue up and leave with the effluent gas, **233**, through the gas/dust/granular mixture exit, **216**, the connecting means, **241**, and into the gas/granular separator means, **230**, where the granules are removed via the granular removal means, **231**. The remaining gas and dust leave through the gas/dust top exit tube, **229**, then enter the gas/dust separation means, **217**, where most of the dust, **225**, is removed from the gas, **233**, and ultimately leaves the system via the dust removal means, **218**, while the gas, **233**, and residual dust leaves via an exit, **219**.

To accomplish the removal of large granules the average top level, **222**, is very close to the gas/dust/granular mixture exit, **216**, and consequently some of the product granules, **236**, thrown up above the bed do not describe arcs as they rise then fall under gravity in the disengaging space, **224**, but continue with the entrained dust, **225**, out the gas/dust/granular mixture exit, **216**. Since the average bed level, **222**, is closer to the exit, **216**, the bed level, **222**, can be taller and/or the overall height, **244**, can be shorter compared to the prior art as shown in Fig 1.

Turning to Fig 2b there is shown in detail the various mechanisms which cause the product granules, **236**, to be carrried out the gas exit, **216**. The basic mechanism is the random ejection of product granules, **236**, from the top of the bed, **222**, and the pneumatic conveying of these granules out the gas/dust/granular exit, **216**. In addition the bed level oscillates up and down due to the formation of gas slugs, **240**, which lift sections of the bed up to the high level, **232**, until they break through and the bed level recedes to the low level, **234**. It is also possible for the bed to reach extra high levels, **235**, where the bed is above the exit briefly. The exit tube, **241**, can be attached to the exit, **216**, at 90° as shown or sloped above or below the horizontal. The angle chosen can be determined by the application of standard pneumatic conveying calculations using the gas velocity in the exit tube, **241**.

Turning now to Fig 3 there is shown a more detailed schematic of a product separator, **330**, with an integrated heat recovery system, **301**, suitable for high temperature and high purity applications. The gas/dust/granular mixture, **333**, enters the product separator, **330**, through an inlet, **357**, which goes through the heat recovery system, **301**, via a penetration, **358**; the gas and dust, **356**, then separate to the top and exit via the exit tube, **329**, while the granules, **336**, separate to the bottom exit, **331**, where it is fluidized by a purge stream, **359**, and withdrawn as needed.

The heat recovery system, **301**, is comprised of a heat transfer fluid, **360**, contained in a container, **351**, which is shaped to capture heat, **350**, from the wall of the product separator and has an inlet, **354**, and an outlet, **355**, for the heat transfer fluid, **360**. The container can use various heat transfer fluids such as water or hot oil. It is usually advantageous for the container to be a pressure vessel to permit heat recovery at higher temperatures. The heat may be transferred from the wall to the container by radiation, conduction or convection and well-known heat transfer techniques can be used to enhance the heat transfer from the gas and solids to the wall. Similarly, well-known gas-solids removal techniques, such as cyclones or filters, can be used to enhance the gas-solids separation.

In a particularly advantageous design, the heat is transferred by radiation from the hot surface of the product separator to a pressurized container which has water, **352**, coming in through the inlet, **354**, and steam, **353**, leaving through the exit, **355**.

An example using Fig 2 would be as follows. The diameter of the container is 300 mm, the overall height of the liner, **244**, is 7 meters, the average bed level, **222**, is 6 meters, the high level is about 6.6 meters and the low level is about 5.4 meters. The gas superficial velocity at the top of the container is 4.7 ft/s (1.4 m/s). The average particle size of the granules is 1mm and the terminal velocity is 21.8 ft/s (6.56 m/s). The particle terminal velocity is thus about 4 times the superficial gas velocity. This means that in order to carry the granules out of the reactor, the local velocity in areas just above the bed must have local surges where it is 4 times higher than average. Velocity surges of this magnitude occur close to the top of the bed at about 20 cm above the bed. The slug, **240**, has a maximum length of about 1.2 meter, and so the periodic growth and bursting of the slug provides the variation in height of 1.2 meters between low and high level. As the slug bursts, it also accelerates the granular particles which are then entrained out of the reactor. Thus the granular removal varies with the pulsing of the slugs, **240**

In comparison, for Fig 1 under similar operating conditions with an average bed level, **122**, of 6 meters, the overall height would be 10 meters in order to allow for the disengaging space normally required under the prior art.

The granules and gas at the bottom of the reactor are at 700 °C, then are heated up and leave the reactor as stream, **233**, via exit, **216**, at a temperature of 800 °C. They enter the cyclonic product separator, **230**, through a tangential inlet which forces the gas and solids to the wall of the vessel to improve gas to wall heat transfer. The diameter of the cyclone is 10 inches (250 mm) and the length is 6 ft (1.8m). This is longer than needed for solely the solids removal in order to provide sufficient surface area for heat transfer. The gas and granules both leave at 600 °C. The dust/gas separator, **217**, is of a similar size but only removes about half the heat because of the reduction in the temperature difference. The gas and dust then leave the dust/gas separator at 500 °C. Both heat recovery systems recover the heat as 150 psig steam, which is a standard utility useful in the facility for a variety of purposes and thus always in demand.

While the invention has been described in connection with a preferred embodiment, it is not intended to limit the scope of the invention to the particular form set forth, but on the contrary, it is intended to cover such alternatives, modifications, and equivalents as may be included within the spirit and scope of the invention as defined by the appended claims.

Throughout this specification and claims which follow, unless the context requires otherwise, the word "comprise", and variations such as "comprises" and "comprising", will be understood to imply the inclusion of a stated integer or step or group of integers or steps but not the exclusion of any other integer or step or group of integers or steps.

The reference in this specification to any prior publication (or information derived from it), or to any matter which is known, is not, and should not be taken as an acknowledgment or admission or any form of suggestion that that prior publication (or information derived from it) or known matter forms part of the common general knowledge in the field of endeavour to which this specification relates.

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:

1. A method of operation of a fluidized bed deposition reactor for top removal of granular material comprising:

providing a vertical generally cylindrical reactor of a predetermined height, said reactor having at least one gas inlet at or near the bottom thereof and at least one gas and solids outlet at or near the top thereof,

providing at least one gas and granular product separator to separate gas from granular product and a conduit between the gas and solids outlet and said at least one separator,

establishing a heated reaction zone;

providing a fluidizing gas to the reaction zone at a predetermined flow rate,

providing granular particles to the reaction zone to establish the desired fluidized bed, said particles having a variable fluidized height, the bed being fluidized to establish a bubbling fluidized bed with a top of a defined stable height, a disengaging space being provided above said top, the height of said disengaging space being no greater than the distance between the defined top of the bubbling bed and the gas and solids outlet of the container,

providing a second gas to the reaction zone while adjusting the flow rate of the fluidizing gas so that a majority of the granular particles are retained in the reactor while maintaining bubbling, the second gas depositing a coating on said granular particles, thus increasing the size and weight of the particles resulting in an increase in the height of the bed, and

allowing the height of the bed to increase and the disengaging height to decrease until the bursting of bubbles near the surface of the bed periodically throws granular particles out of the gas and solids outlet of the reactor, along the conduit and into the at least one gas and granular product separator,

wherein the flow rates of the fluidizing gas and the second gas are controlled so as to establishing a stable height of the bed.

2. The method of claim 1 further including recoverying heat while separating the granular product from the gas comprising using the at least one gas and granular product separator in combination with one or more heat recovery systems.

3. The method of claim 2 wherein at least one of the heat recovery systems recovers heat by radiation to a heat recovery boiler.

4. The method of any one of claims 1 to 3, wherein at least one means for granular product removal is also provided at the bottom of the reactor.

5. The method of any one of claims 1 to 4, wherein the at least one gas and granular product separator provides one or more product streams comprising particles of respective different average particle sizes.

6. A method of operation of a fluidized bed deposition reactor for top removal of granular material, substantially as hereinbefore described with reference to the accompanying drawings.

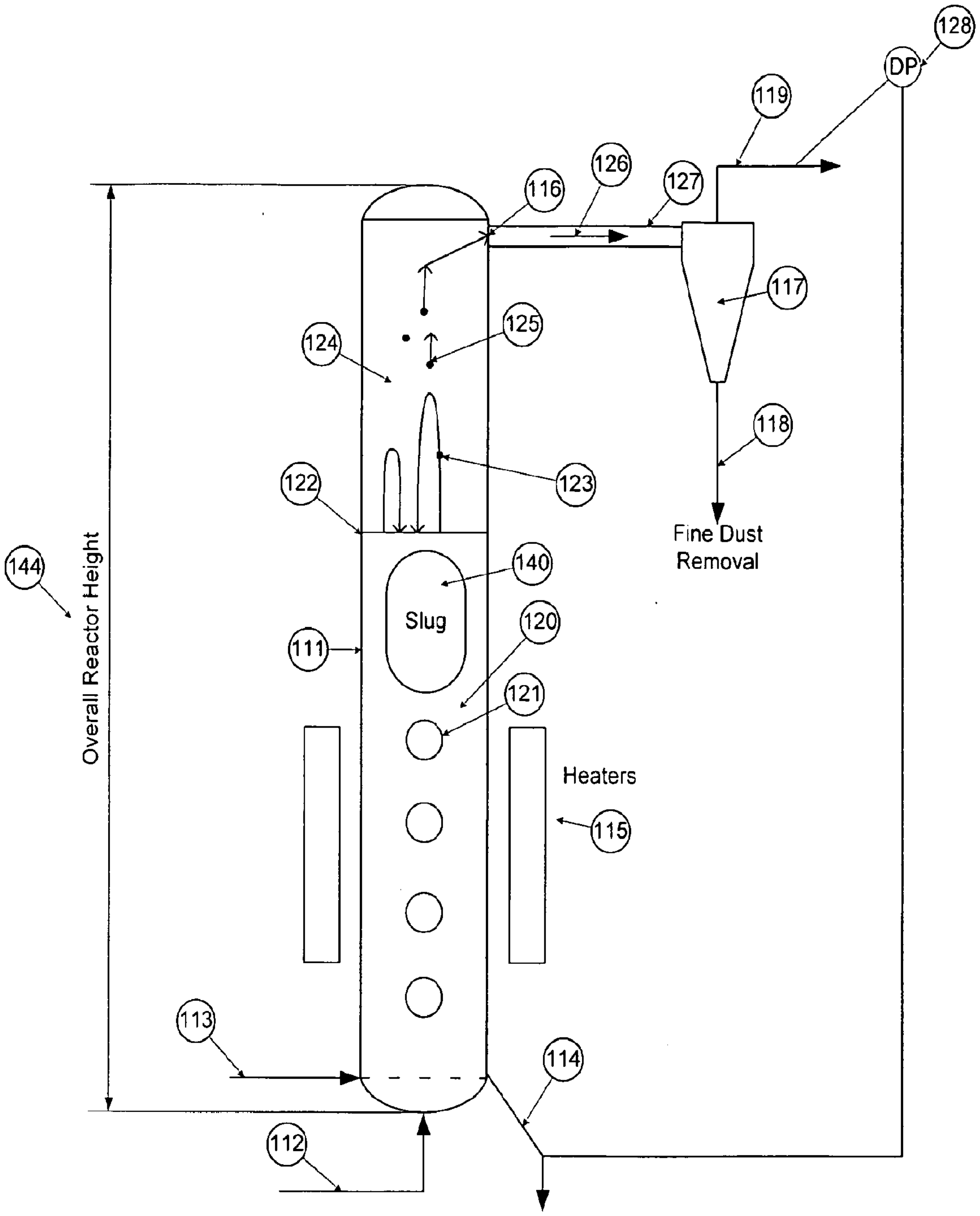

Fig 1. Typical Fluidized Bed Deposition Reactor

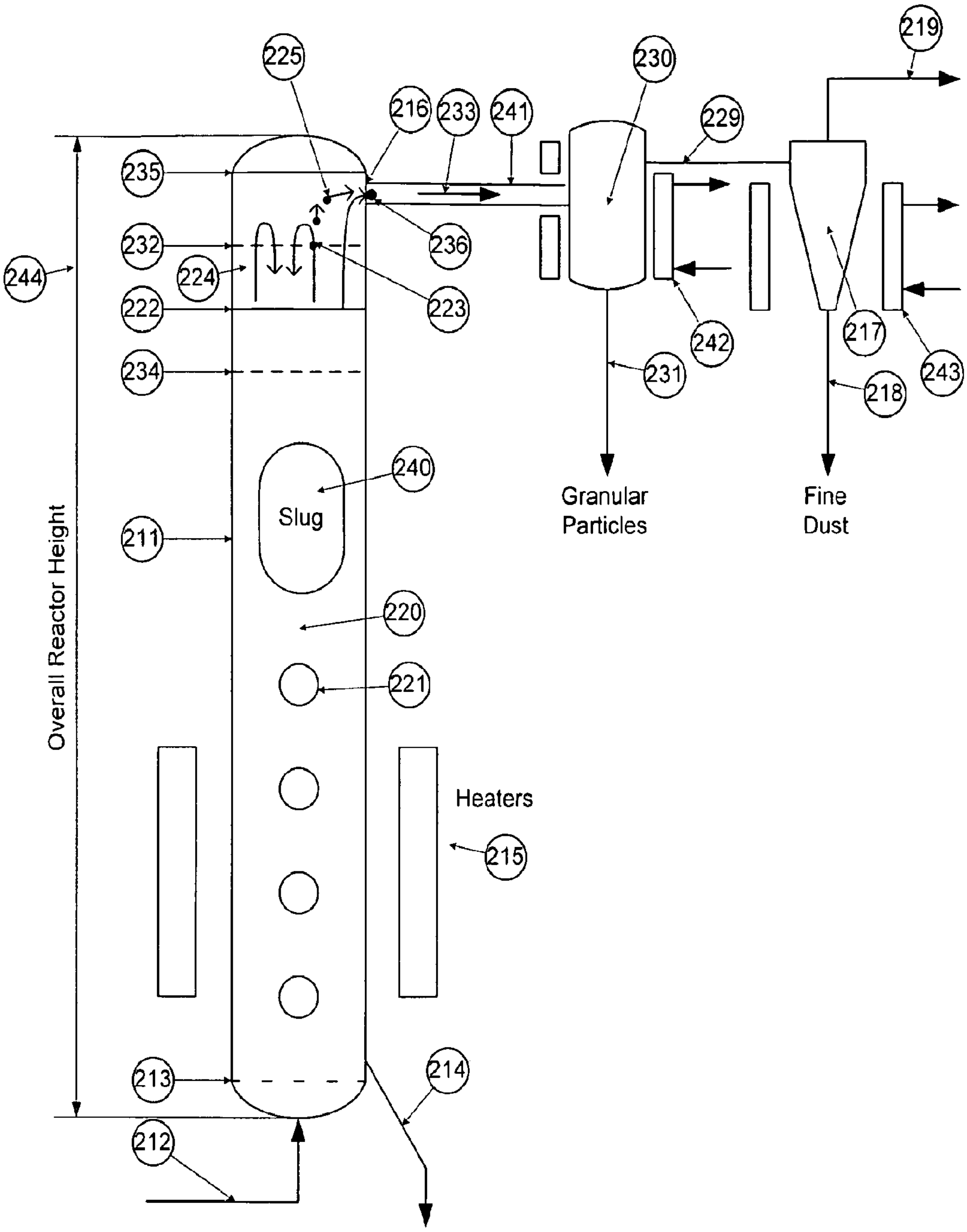

Fig 2a. The Invention

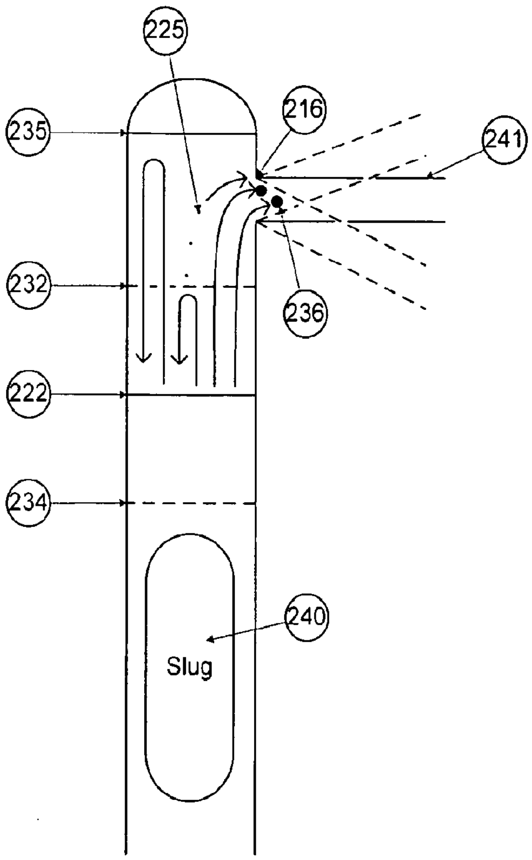

Fig 2b. Detail schematic of the granular particle removal mechanism

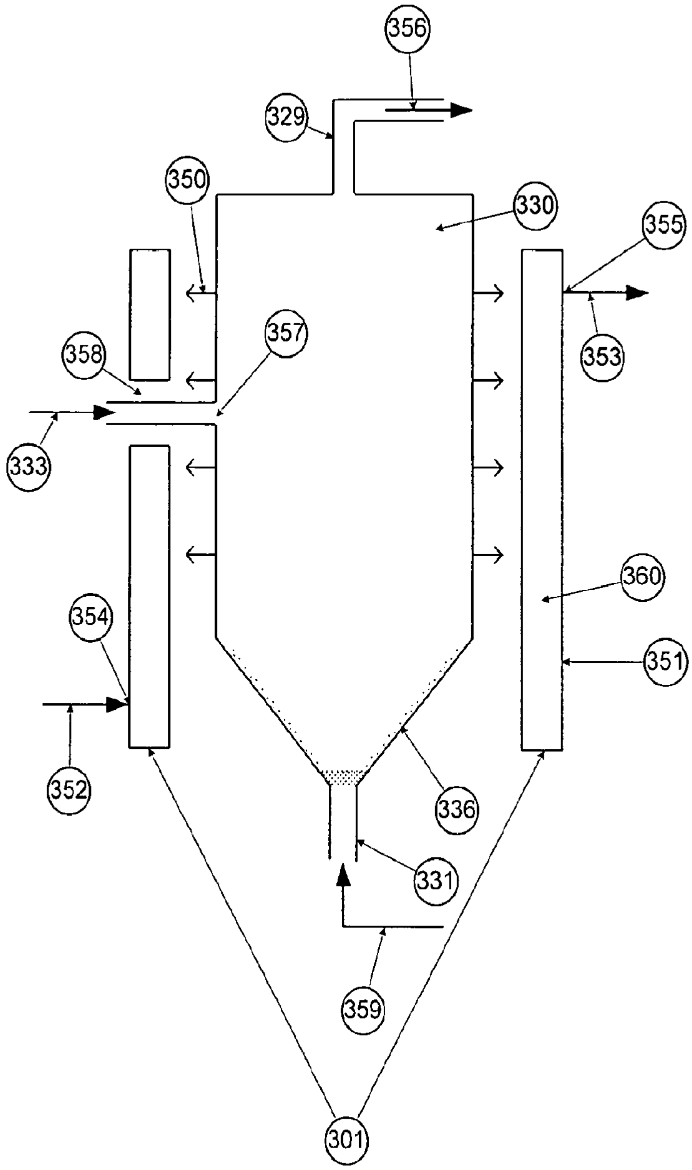

Fig 3. Schematic of the product separator with integrated heat recovery