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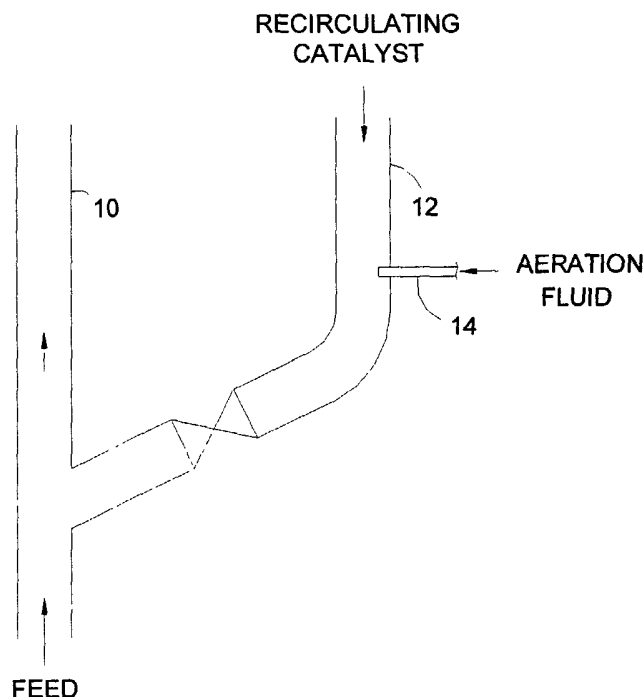
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(54) Title: CONTROLLING SOLIDS CIRCULATION IN A GAS-SOLIDS REACTION SYSTEM



(57) Abstract: This invention provides a method for controlling solids circulation in a gas-solids reaction system. The method entails aerating solid particles in a standpipe. Aeration fluid is injected into the standpipe at the appropriate location to increase apparent density of the solid particles.

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CONTROLLING SOLIDS CIRCULATION IN A GAS-SOLIDS REACTION SYSTEM

[0001] This application claims the benefit of U.S. Provisional Application No. 60/390,486, filed June 21, 2002, the contents of which are fully incorporated herein by reference.

Field of the Invention

[0002] This invention relates to controlling solids circulation in a gas-solids reaction system. In particular, this invention relates to controlling solids circulation in reaction systems employing solid particle catalysts.

Background of the Invention

[0003] Conventional gas-solids reaction systems are continuous systems which contact feed with a solid catalyst, and circulate the solid catalyst for additional contact with fresh feed. The circulated catalyst typically passes through a standpipe type of arrangement prior to contact with fresh feed. The standpipe is typically connected to a valve type of arrangement to control the amount of catalyst that is circulated to contact the feed.

[0004] For example, U.S. Patent No. 6,165,353 discloses a gas solids reaction system in which solid catalyst is collected in a blend vessel at the top of a reactor. The catalyst passes through a standpipe, with the flow of the catalyst being controlled by a slide valve. Following the slide valve, a pressurization fluid is used to provide a fluidizing function to maintain flow of the catalyst through a discharge point where contact is made with feed.

[0005] U.S. Patent No. 6,143,253 discloses a gas solids reaction system in which catalyst is circulated through a standpipe. Flow from the standpipe is controlled by a slide valve. After the catalyst passes through the slide valve, it passes to a conduit section. In the conduit section, a nozzle injects steam to control the density of the catalyst.

[0006] U.S. Patent No. 5,554,341 discloses a fluidized catalytic unit which has a riser-reactor type unit connected to a regenerator. Solid catalyst is withdrawn from the regenerator through a generally vertically oriented standpipe

and the flow rate is controlled as desired by one or more slide valves positioned in the standpipe. The catalyst then flows through a J-bend transfer line wherein catalyst flow is converted from a principally downward to principally upward direction. The catalyst then enters the feed injection cone and associated mixing zone. Catalyst transfer through the standpipe or the transfer line and the J-bend transfer line can be facilitated by injecting gas, preferably steam, into the catalyst bed. The catalyst bed is transported by gas flow, preferably steam, to the feed injection cone assembly whereupon the catalyst is contacted with oil and a gas, preferably steam.

[0007] Bodin et al., in *Powder Technology*, 124 (2002), 8-17, indicate that the role of the standpipe is to transfer solids from a low pressure vessel to a high-pressure vessel, and that aeration along the length of the standpipe is necessary to keep the solids fluidized and prevent irregular flow or low pressure buildup. The authors further indicate that startup operation is particularly difficult, because solids flow rate is lower than normal. Therefore, proper aeration during startup is particularly important so as to attain stable solids flow.

[0008] In gas-solids systems in general, it is important to properly control the solid catalyst being circulated through the standpipe in order to gain the maximum benefit at point of feed contact. Accordingly, additional methods of controlling solids circulation in gas-solids reaction systems are highly desired. With better control, it is possible to increase the amount of feed that can be introduced into a gas-solids reaction system without increasing the amount of catalyst already in the system.

Summary of the Invention

[0009] This invention provides a gas-solids reactor system having improved solids circulation. One way in which this is accomplished is by increasing the apparent density of the solid that is being circulated, and/or reducing pressure fluctuation through the standpipe. Increasing the apparent density can also affect the distribution of catalyst inventory in the system, e.g., between the standpipe and other equipment in fluid solid communication with the standpipe. Another benefit is to affect the pressure balance around the standpipe and other equipment in which fluid solid communication occurs.

[0010] In one embodiment, this invention provides a method of aerating solid particles in a standpipe, which comprises providing a standpipe having an internal wall and solid particles within the standpipe. Aeration fluid is injected at a point away from the internal wall, wherein at least about 25%, preferable at least about 50% of the aeration fluid is injected at a point away from the internal wall, so as to aerate the solid particles in the standpipe.

[0011] In another embodiment, the invention provides a method of aerating solid catalyst particles in a standpipe, which comprises providing a standpipe having a centerline, an internal wall and solid particles within the standpipe. Aeration fluid is injected into the standpipe at a point such that at least a portion of the fluid is injected at a location $r/R < 0.75$, wherein r is defined as the radial distance from the centerline of the standpipe to the point of aeration fluid injection, and R is defined as the radial distance from the centerline of the standpipe to the internal wall of the standpipe, so as to aerate the solid particles in the standpipe.

[0012] In yet another embodiment, the invention comprises a method of aerating solid particles in a standpipe, which comprises providing a standpipe having an internal wall and solid particles within the standpipe. Aeration fluid is injected at a point away from the internal wall such that the solid particles are aerated in the standpipe and have an apparent density of at least about 50% of their maximum apparent density.

[0013] The invention also provides a method of aerating solid particles in a standpipe of a gas-solids reactor system in which aeration fluid is injected through at least two orifices, more preferably through at least three orifices, into the standpipe in inwardly radial directions, with the orifices being located at different azimuthal positions on the standpipe. In one embodiment, at least one of the orifices is positioned to flow aeration fluid in the azimuthal direction around a circumference of the internal wall. Preferably, the aeration orifices are separated at a distance of from $\pi/5$ to π radians, more preferably at a distance of from $\pi/4$ to $2\pi/3$ radians.

[0014] In another embodiment of the invention, injection of aeration fluid internal to the standpipe and at an azimuthal position at the internal wall occur at

the same time. For example, in one embodiment, the aeration fluid is injected at a point away from the internal wall of the standpipe, preferably through a sparger, and additional aeration fluid is injected through at least one orifice in at an azimuthal position at the internal wall.

[0015] The standpipe can be in any conventional gas-solids reaction systems. Examples include oxygenate to olefin reaction processes, fluidized catalytic cracking reaction processes, processes for oxidation of n-butane to maleic anhydride, and processes for ammonia oxidation of propylene or propane to acrylonitrile. In one embodiment, the solid particles in the standpipe have an apparent density of at least about 20 lb/ft³.

Brief Description of the Drawings

[0016] Examples of various embodiments of this invention are shown in the attached Drawings, wherein:

[0017] Fig. 1 shows a standpipe connected to a riser-reactor, with aeration fluid being input into the standpipe prior to the catalyst passing across a slide valve;

[0018] Fig. 2 shows a standpipe arrangement that includes multiple standpipe portions;

[0019] Fig. 3 shows a single standpipe arrangement having microporous gas spargers;

[0020] Fig. 4 shows a horizontal section view of a standpipe having a sparger and two orifices for injecting aeration fluid;

[0021] Fig. 5 shows a graph of dynamic differential pressure (DP) measurements taken at multiple locations in a standpipe with aeration fluid injected through one orifice;

[0022] Fig. 6 shows a graph of dynamic differential pressure (DP) measurements taken at multiple locations in a standpipe with aeration fluid injected through two orifices;

[0023] Fig. 7 shows a graph of dynamic differential pressure (DP) measurements taken at multiple locations in a standpipe with aeration fluid injected through three orifices; and

[0024] Fig. 8 shows a graph of dynamic differential pressure (DP) measurements taken at multiple locations in a standpipe with aeration fluid injected through a sparger and two orifices.

Detailed Description of the Invention

I. Increasing Catalyst Apparent Density and Reducing Pressure Fluctuations

[0025] Additional feed can be input into a gas-solids reaction system by increasing the amount of solid catalyst that is circulated in the system. In conventional gas-solids reaction systems, catalysts are reused to contact additional feed. Typically, the catalyst is regenerated before circulating; however, catalyst can be reused without regenerating by circulating through a separate line. In either event, the solid catalyst is typically circulated through a standpipe type of arrangement, before contacting additional feed. In general, the more catalyst that is circulated through this standpipe arrangement, the more feed that can be input into the reactor. This invention allows for increased circulation without having to add more catalyst or make further adjustments to the catalyst regenerator.

[0026] According to this invention, the amount of catalyst that is circulated in the system is affected by increasing the apparent density of the catalyst, while reducing pressure fluctuations within the standpipe. In one embodiment, the apparent density can actually be increased, and/or pressure fluctuations reduced, by appropriately aerating the catalyst being circulated. Preferably, aeration is applied at the standpipe region of the gas-solids reaction system. The standpipe region is generally the region where solids is returned to the reactor portion of the system against a pressure gradient. Conventionally, a valve such as a slide valve is used to regulate the amount of catalyst circulation in the system. In such cases, the standpipe will be the region before the pressure let-down across such a valve. Following the standpipe and the pressure let-down valve will generally be a conduit that transports the solid particles into the reactor portion of the system.

[0027] In general, this invention provides a way to aerate circulated solid catalyst to approach maximum apparent density, expressed as pressure drop per unit length. In this invention, maximum apparent density is the density of the

catalyst at a point in which incipient fluidization occurs on a fluidization curve. See, *Circulating Fluidized Beds*, J.R. Grace, A.A. Avidan, and T.M Knowlton, Eds., Blackie Academic & Professional New York (1997). The point of incipient fluidization of solid catalyst particles is the lowest gas velocity at which all of the particles are suspended by the fluidizing gas. At this condition, the pressure drop generated by the fluidizing gas is equal to the weight of solid catalyst particles per unit area. Although, the absolute value of the maximum apparent density varies according to the actual density of the particular particles being fluidized and the sizes of the particles, the maximum apparent density of any fluidized solids can be readily determined by those of skill in the art. One way of measuring apparent density is by determining the difference in the pressures, measured at two elevations along the standpipe, divided by the height between the pressure measurements.

[0028] Desirably, the catalyst in the standpipe region of the gas-solids reaction system of this invention has an apparent density of at least about 50% of the maximum apparent density of the catalyst. Preferably, the catalyst in the standpipe region of the gas-solids reaction system of this invention has an apparent density of at least about 60% of the maximum apparent density of the catalyst; more preferably, at least about 80% of the maximum apparent density of the catalyst.

[0029] In one embodiment of the invention, the apparent density of the catalyst in the standpipe is at least about 20 lb/ft³, and up to as high as the maximum apparent density of the solid catalyst in the standpipe. Preferably, the apparent density of the catalyst in the standpipe is at least about 25 lb/ft³, more preferably at least about 30 lb/ft³, and up to as much as about 60 lb/ft³.

[0030] In this invention, the catalyst that is circulated at increased apparent density flows through a standpipe arrangement, with reduced fluctuations in differential pressure across the standpipe. In one embodiment, the catalyst flows through the standpipe and encounters feed input into the reactor system. In another embodiment, the standpipe is connected to a region of the reactor where feed is in the gas phase and flowing at a rate that results in the catalyst flowing faster through the reactor portion of the system than through the standpipe. In a

particular embodiment, the solid catalyst flows through a riser portion of the reactor.

[0031] Desirably, the solid catalyst flows through the standpipe location at a velocity of about 0.25 m/sec to about 5 m/sec. Preferably, the solid catalyst flows through the standpipe location at a velocity of from about 0.75 m/sec to about 4 m/sec; more preferably, from about 1 m/sec to about 3 m/sec.

[0032] In certain embodiments of the invention, the solid catalyst flows from the standpipe location through a conduit and into the reactor portion of the system. In this embodiment, it is desirable that the solid catalyst flows through the reactor portion of the system at a velocity of from about 1 m/sec to about 30 m/sec; preferably, from about 3 m/sec to about 25 m/sec; more preferably, from about 5 m/sec to about 20 m/sec.

[0033] Aeration of the catalyst in the standpipe location should be such that formation of gas bubbles is minimized. One way to accomplish this is to inject aeration fluid into the standpipe in such a manner so as to increase apparent density of the solid material being circulated through the standpipe. Injection should be such that the aeration fluid is injected into the standpipe to obtain good dispersion of fluid within the standpipe.

[0034] In one embodiment, aeration fluid is injected at a point away from an internal wall of the standpipe, with the internal wall being the inner surface of the wall that is in contact with the gas and solid particles. Preferably, at least about 25%, and more preferably at least about 50% of the aeration fluid is injected at a point away from the internal wall; more preferably, at least about 75% of the aeration fluid is injected at a point away from the internal wall; and most preferably, 100% of the aeration fluid is injected at a point away from the internal wall.

[0035] In another embodiment of the invention, aeration fluid is injected into the standpipe at a point such that at least a portion of the fluid is injected at a location $r/R < 0.75$, wherein r is the radial distance from the centerline of the standpipe to the point of aeration fluid injection, and R is the radial distance from the centerline of the standpipe to the internal wall of the standpipe. Preferably, the

fluid is injected at a location $r/R < 0.6$; more preferably the fluid is injected at a location $r/R < 0.5$.

[0036] It is also desirable to disperse the aeration fluid substantially uniformly within the standpipe. For example, this can be achieved by introducing the aerating gas using a series of orifices, the orifices being separated at a distance of from about $\pi/5$ to about π radians. Preferably the orifices are separated at a distance of from about $\pi/4$ to about $2\pi/3$ radians. Examples of preferred embodiments include the use of 2 orifices spaced about 180 degrees from one another; the use of 3 orifices spaced about 120 degrees from one another; and the use of 4 orifices spaced about 90 degrees from one another.

[0037] It is particularly desirable to have at least two orifices located substantially in the same horizontal plane. Preferably, there are at least two orifices located within about 1/2 foot of a horizontal baseline through the standpipe, more preferably within about 1/3 foot, and most preferably within about 1/4 foot. For example, in an embodiment when two orifices are located substantially in the same horizontal plane, it is preferred that the two orifices be within 1/2 foot of a horizontal baseline, with the two orifices located at about 180 degrees apart in the horizontal plane.

[0038] In another embodiment, the aeration fluid is injected through at least two orifices into the standpipe in inwardly radial directions, with the orifices being located at different azimuthal positions on the standpipe; meaning that the orifices are located at different positions along the circumference of the standpipe. In a particular embodiment, at least one of the orifices is positioned to flow aeration fluid in the azimuthal direction around a circumference of the internal wall. In another embodiment, aeration fluid is injected through a sparger or pipe-type of device, which has at least one orifice. The sparger is positioned so that it projects within the standpipe. Orifices on the sparger are positioned to flow aeration fluid in an inward direction. In one embodiment, the orifices are positioned to flow aeration fluid in an azimuthal direction; in another embodiment, in a direction against flow of solid particles; and in another embodiment, in a direction with the flow of solid particles.

[0039] In a particular embodiment, the aeration fluid is injected radially through at least three orifices, with each orifice being located at different azimuthal locations around the circumference of the internal wall of the standpipe. Desirably, the aeration fluid is injected inwardly of the standpipe. Preferably, at least one orifice is positioned to inject aeration fluid substantially uniformly in the azimuthal direction. Preferably, in the embodiment where at least three orifices are used, and preferably up to 8 or 10 orifices, the aeration orifices are separated at a distance of from $\pi/5$ to $7\pi/8$ radians, more preferably at a distance of from $\pi/4$ to $2\pi/3$ radians from one another.

[0040] According to this invention, aeration fluid is injected either internal to the standpipe or in the azimuthal direction to increase the apparent density of solid particles within the standpipe. In a preferred embodiment, the standpipe is configured to inject aeration fluid internal to the standpipe as well as in the azimuthal direction. For example, in one embodiment, the aeration fluid is injected at a point away from the internal wall of the standpipe, and additional aeration fluid is injected through orifices in an azimuthal direction around a circumference of the internal wall.

[0041] In one embodiment the devices for introducing the aerating gases can be a pipe with appropriate orifices. The pipe can be straight or shaped, such as in the form of an arc, cross or ring.

[0042] Aeration fluid is applied in a standpipe to offset the effects of compression. Without aeration, solid particles in the standpipe will defluidized, and flow of the solid particles across any pressure gradient point, such as across a slide valve, will be severely impeded. With too much aeration, however, the flow of solid particles across a pressure gradient will also be impeded.

[0043] The rate of injection of aeration fluid will depend upon many factors such as how well the aeration fluid is dispersed, how well the fluid permeates the solids, the particle density, and the particle size distribution. In general, the easier it is for the fluid to permeate the solids, the lower the rate of injection of aeration fluid that is needed. Additionally, with respect to the particle density, the greater the particle density of the solid material that is being aerated, the greater the rate of injection of aeration fluid is needed.

[0044] The rate of injection of aeration fluid into the standpipe should be effective to increase apparent density of the solids in the standpipe. Desirably, the aeration fluid is injected at a velocity of at least about 0.005 ft/sec. According to this invention, the velocity of the aeration fluid is the volumetric flow rate of the aeration fluid divided by the cross sectional area of the standpipe.

[0045] The velocity aeration fluid into the standpipe should also be such that there is no substantial upflow or elutriation of the solid particles, since the normal operation of the standpipe is to establish flow of solid particles against gravity. Preferably, the velocity of the aeration fluid injected into the standpipe is from about 0.005 ft/sec to about 3 ft/sec, more preferably from about 0.01 ft/sec to about 2.5 ft/sec.

[0046] In one embodiment of the invention the standpipe has an internal diameter of at least about 2 feet, 2½ feet or 3 feet. In another embodiment, the standpipe can be larger in internal diameter, with diameters of at least about 4 feet or 4½ feet, and up to 5 or 6 feet.

II. Solid Particles that Can be Used

[0047] This invention can be used with any type of particle that is capable of being flowed in a conventional fluid state. In one embodiment, the solid particles are Geldart group A particles. See, Geldart, A., "Types of Gas Fluidization" *Powder Technol.* 7 (1973) 285-292, which is fully incorporated herein by reference.

[0048] In one embodiment, the solid catalyst particles which are circulated through the gas-solids reactor system of this invention are molecular sieve catalysts. Any conventional molecular sieve can be used. Examples include zeolite as well as non-zeolite molecular sieves, and are of the large, medium or small pore type. Non-limiting examples of these molecular sieves are the small pore molecular sieves, AEI, AFT, APC, ATN, ATT, ATV, AWW, BIK, CAS, CHA, CHI, DAC, DDR, EDI, ERI, GOO, KFI, LEV, LOV, LTA, MON, PAU, PHI, RHO, ROG, THO, and substituted forms thereof; the medium pore molecular sieves, AFO, AEL, EUO, HEU, FER, MEL, MFI, MTW, MTT, TON, and substituted forms thereof; and the large pore molecular sieves, EMT, FAU, and substituted forms thereof. Other molecular sieves include ANA, BEA, CFI, CLO,

DON, GIS, LTL, MER, MOR, MWW and SOD. Non-limiting examples of the preferred molecular sieves, particularly for converting an oxygenate containing feedstock into olefin(s), include AEL, AFY, BEA, CHA, EDI, FAU, FER, GIS, LTA, LTL, MER, MFI, MOR, MTT, MWW, TAM and TON. In one preferred embodiment, the molecular sieve of the invention has an AEI topology or a CHA topology, or a combination thereof, most preferably a CHA topology.

[0049] Molecular sieve materials all have 3-dimensional, four-connected framework structure of corner-sharing TO_4 tetrahedra, where T is any tetrahedrally coordinated cation. These molecular sieves are typically described in terms of the size of the ring that defines a pore, where the size is based on the number of T atoms in the ring. Other framework-type characteristics include the arrangement of rings that form a cage, and when present, the dimension of channels, and the spaces between the cages. See van Bekkum, et al., *Introduction to Zeolite Science and Practice, Second Completely Revised and Expanded Edition*, Volume 137, pages 1-67, Elsevier Science, B.V., Amsterdam, Netherlands (2001).

[0050] The small, medium and large pore molecular sieves have from a 4-ring to a 12-ring or greater framework-type. In a preferred embodiment, the molecular sieves have 8-, 10- or 12- ring structures or larger and an average pore size in the range of from about $3\text{\AA}$ to $15\text{\AA}$. In the most preferred embodiment, the molecular sieves of the invention, preferably silicoaluminophosphate molecular sieves, have 8- rings and an average pore size less than about $5\text{\AA}$, preferably in the range of from $3\text{\AA}$ to about $5\text{\AA}$, more preferably from $3\text{\AA}$ to about $4.5\text{\AA}$, and most preferably from $3.5\text{\AA}$ to about $4.2\text{\AA}$.

[0051] Molecular sieves, particularly zeolitic and zeolitic-type molecular sieves, preferably have a molecular framework of one, preferably two or more corner-sharing $[\text{TO}_4]$ tetrahedral units, more preferably, two or more $[\text{SiO}_4]$, $[\text{AlO}_4]$ and/or $[\text{PO}_4]$ tetrahedral units, and most preferably $[\text{SiO}_4]$, $[\text{AlO}_4]$ and $[\text{PO}_4]$ tetrahedral units. These silicon, aluminum, and phosphorous based molecular sieves and metal containing silicon, aluminum and phosphorous based molecular sieves have been described in detail in numerous publications including for example, U.S. Patent No. 4,567,029 (MeAPO where Me is Mg, Mn, Zn, or

Co), U.S. Patent No. 4,440,871 (SAPO), European Patent Application EP-A-0 159 624 (ELAPSO where El is As, Be, B, Cr, Co, Ga, Ge, Fe, Li, Mg, Mn, Ti or Zn), U.S. Patent No. 4,554,143 (FeAPO), U.S. Patents No. 4,822,478, 4,683,217, 4,744,885 (FeAPSO), EP-A-0 158 975 and U.S. Patent No. 4,935,216 (ZnAPSO, EP-A-0 161 489 (CoAPSO), EP-A-0 158 976 (ELAPO, where EL is Co, Fe, Mg, Mn, Ti or Zn), U.S. Patent No. 4,310,440 (AlPO₄), EP-A-0 158 350 (SENAPSO), U.S. Patent No. 4,973,460 (LiAPSO), U.S. Patent No. 4,789,535 (LiAPO), U.S. Patent No. 4,992,250 (GeAPSO), U.S. Patent No. 4,888,167 (GeAPO), U.S. Patent No. 5,057,295 (BAPSO), U.S. Patent No. 4,738,837 (CrAPSO), U.S. Patents Nos. 4,759,919, and 4,851,106 (CrAPO), U.S. Patents Nos. 4,758,419, 4,882,038, 5,434,326 and 5,478,787 (MgAPSO), U.S. Patent No. 4,554,143 (FeAPO), U.S. Patent No. 4,894,213 (AsAPSO), U.S. Patent No. 4,913,888 (AsAPO), U.S. Patents Nos. 4,686,092, 4,846,956 and 4,793,833 (MnAPSO), U.S. Patents Nos. 5,345,011 and 6,156,931 (MnAPO), U.S. Patent No. 4,737,353 (BeAPSO), U.S. Patent No. 4,940,570 (BeAPO), U.S. Patents Nos. 4,801,309, 4,684,617 and 4,880,520 (TiAPSO), U.S. Patents Nos. 4,500,651, 4,551,236 and 4,605,492 (TiAPO), U.S. Patents No. 4,824,554, 4,744,970 (CoAPSO), U.S. Patent No. 4,735,806 (GaAPSO) EP-A-0 293 937 (QAPSO, where Q is framework oxide unit [QO₂]), as well as U.S. Patents Nos. 4,567,029, 4,686,093, 4,781,814, 4,793,984, 4,801,364, 4,853,197, 4,917,876, 4,952,384, 4,956,164, 4,956,165, 4,973,785, 5,241,093, 5,493,066 and 5,675,050, all of which are herein fully incorporated by reference.

[0052] Other molecular sieves include those described in EP-0 888 187 B1 (microporous crystalline metallophosphates, SAPO₄ (UIO-6)), U.S. Patent No. 6,004,898 (molecular sieve and an alkaline earth metal), U.S. Patent Application Serial No. 09/511,943 filed February 24, 2000 (integrated hydrocarbon co-catalyst), PCT WO 01/64340 published September 7, 2001 (thorium containing molecular sieve), and R. Szostak, *Handbook of Molecular Sieves*, Van Nostrand Reinhold, New York, New York (1992), which are all herein fully incorporated by reference.

[0053] The more preferred silicon, aluminum and/or phosphorous containing molecular sieves, and aluminum, phosphorous, and optionally silicon,

containing molecular sieves include aluminophosphate (ALPO) molecular sieves and silicoaluminophosphate (SAPO) molecular sieves and substituted, preferably metal substituted, ALPO and SAPO molecular sieves. The most preferred molecular sieves are SAPO molecular sieves, and metal substituted SAPO molecular sieves. In an embodiment, the metal is an alkali metal of Group IA of the Periodic Table of Elements, an alkaline earth metal of Group IIA of the Periodic Table of Elements, a rare earth metal of Group IIIB, including the Lanthanides: lanthanum, cerium, praseodymium, neodymium, samarium, europium, gadolinium, terbium, dysprosium, holmium, erbium, thulium, ytterbium and lutetium; and scandium or yttrium of the Periodic Table of Elements, a transition metal of Groups IVB, VB, VIB, VIIB, VIIIB, and IB of the Periodic Table of Elements, or mixtures of any of these metal species. In one preferred embodiment, the metal is selected from the group consisting of Co, Cr, Cu, Fe, Ga, Ge, Mg, Mn, Ni, Sn, Ti, Zn and Zr, and mixtures thereof. In another preferred embodiment, these metal atoms discussed above are inserted into the framework of a molecular sieve through a tetrahedral unit, such as $[\text{MeO}_2]$, and carry a net charge depending on the valence state of the metal substituent. For example, in one embodiment, when the metal substituent has a valence state of +2, +3, +4, +5, or +6, the net charge of the tetrahedral unit is between -2 and +2.

[0054] In one embodiment, the molecular sieve, as described in many of the U.S. Patents mentioned above, is represented by the empirical formula, on an anhydrous basis:



wherein R represents at least one templating agent, preferably an organic templating agent; m is the number of moles of R per mole of $(M_xAl_yP_z)O_2$ and m has a value from 0 to 1, preferably 0 to 0.5, and most preferably from 0 to 0.3; x, y, and z represent the mole fraction of Al, P and M as tetrahedral oxides, where M is a metal selected from one of Group IA, IIA, IB, IIIB, IVB, VB, VIB, VIIB, VIIIB and Lanthanide's of the Periodic Table of Elements, preferably M is selected from one of the group consisting of Co, Cr, Cu, Fe, Ga, Ge, Mg, Mn, Ni, Sn, Ti, Zn and Zr. In an embodiment, m is greater than or equal to 0.2, and x, y and z are greater than or equal to 0.01.

[0055] In another embodiment, m is greater than 0.1 to about 1, x is greater than 0 to about 0.25, y is in the range of from 0.4 to 0.5, and z is in the range of from 0.25 to 0.5, more preferably m is from 0.15 to 0.7, x is from 0.01 to 0.2, y is from 0.4 to 0.5, and z is from 0.3 to 0.5.

[0056] Non-limiting examples of SAPO and ALPO molecular sieves used in the invention include one or a combination of SAPO-5, SAPO-8, SAPO-11, SAPO-16, SAPO-17, SAPO-18, SAPO-20, SAPO-31, SAPO-34, SAPO-35, SAPO-36, SAPO-37, SAPO-40, SAPO-41, SAPO-42, SAPO-44 (U.S. Patent No. 6,162,415), SAPO-47, SAPO-56, ALPO-5, ALPO-11, ALPO-18, ALPO-31, ALPO-34, ALPO-36, ALPO-37, ALPO-46, and metal containing molecular sieves thereof. The more preferred zeolite-type molecular sieves include one or a combination of SAPO-18, SAPO-34, SAPO-35, SAPO-44, SAPO-56, ALPO-18 and ALPO-34, even more preferably one or a combination of SAPO-18, SAPO-34, ALPO-34 and ALPO-18, and metal containing molecular sieves thereof, and most preferably one or a combination of SAPO-34 and ALPO-18, and metal containing molecular sieves thereof.

[0057] In an embodiment, the molecular sieve is an intergrowth material having two or more distinct phases of crystalline structures within one molecular sieve composition. In particular, intergrowth molecular sieves are described in the U.S. Patent Application Serial No. 09/924,016 filed August 7, 2001 and PCT WO 98/15496 published April 16, 1998, both of which are herein fully incorporated by reference. In another embodiment, the molecular sieve comprises at least one intergrown phase of AEI and CHA framework-types. For example, SAPO-18, ALPO-18 and RUW-18 have an AEI framework-type, and SAPO-34 has a CHA framework-type.

[0058] In one embodiment, the molecular sieves used in the invention are combined with one or more other molecular sieves. In another embodiment, the preferred silicoaluminophosphate or aluminophosphate molecular sieves, or a combination thereof, are combined with one more of the following non-limiting examples of molecular sieves described in the following: Beta (U.S. Patent No. 3,308,069), ZSM-5 (U.S. Patent Nos. 3,702,886, 4,797,267 and 5,783,321), ZSM-11 (U.S. Patent No. 3,709,979), ZSM-12 (U.S. Patent No. 3,832,449), ZSM-12

and ZSM-38 (U.S. Patent No. 3,948,758), ZSM-22 (U.S. Patent No. 5,336,478), ZSM-23 (U.S. Patent No. 4,076,842), ZSM-34 (U.S. Patent No. 4,086,186), ZSM-35 (U.S. Patent No. 4,016,245), ZSM-48 (U.S. Patent No. 4,397,827), ZSM-58 (U.S. Patent No. 4,698,217), MCM-1 (U.S. Patent No. 4,639,358), MCM-2 (U.S. Patent No. 4,673,559), MCM-3 (U.S. Patent No. 4,632,811), MCM-4 (U.S. Patent No. 4,664,897), MCM-5 (U.S. Patent No. 4,639,357), MCM-9 (U.S. Patent No. 4,880,611), MCM-10 (U.S. Patent No. 4,623,527), MCM-14 (U.S. Patent No. 4,619,818), MCM-22 (U.S. Patent No. 4,954,325), MCM-41 (U.S. Patent No. 5,098,684), M-41S (U.S. Patent No. 5,102,643), MCM-48 (U.S. Patent No. 5,198,203), MCM-49 (U.S. Patent No. 5,236,575), MCM-56 (U.S. Patent No. 5,362,697), ALPO-11 (U.S. Patent No. 4,310,440), titanium aluminosilicates (TASO), TASO-45 (EP-A- 0 229,- 295), boron silicates (U.S. Patent No. 4,254,297), titanium aluminophosphates (TAPO) (U.S. Patent No. 4,500,651), mixtures of ZSM-5 and ZSM-11 (U.S. Patent No. 4,229,424), ECR-18 (U.S. Patent No. 5,278,345), SAPO-34 bound ALPO-5 (U.S. Patent No. 5,972,203), PCT WO 98/57743 published December 23, 1988 (molecular sieve and Fischer-Tropsch), U.S. Patent No. 6,300,535 (MFI-bound zeolites), and mesoporous molecular sieves (U.S. Patent Nos. 6,284,696, 5,098,684, 5,102,643 and 5,108,725), which are all herein fully incorporated by reference.

[0059] The molecular sieves are made or formulated into catalysts by combining the synthesized molecular sieves with a binder and/or a matrix material to form a molecular sieve catalyst composition or a formulated molecular sieve catalyst composition. This formulated molecular sieve catalyst composition is formed into useful shape and sized particles by conventional techniques such as spray drying, pelletizing, extrusion, and the like.

[0060] There are many different binders that are useful in forming the molecular sieve catalyst composition. Non-limiting examples of binders that are useful alone or in combination include various types of hydrated alumina, silicas, and/or other inorganic oxide sol. One preferred alumina containing sol is aluminum chlorhydrol. The inorganic oxide sol acts like glue binding the synthesized molecular sieves and other materials such as the matrix together, particularly after thermal treatment. Upon heating, the inorganic oxide sol,

preferably having a low viscosity, is converted into an inorganic oxide matrix component. For example, an alumina sol will convert to an aluminum oxide matrix following heat treatment.

[0061] Aluminum chlorhydrol, a hydroxylated aluminum based sol containing a chloride counter ion, has the general formula of $Al_mO_n(OH)_oCl_p \cdot x(H_2O)$ wherein m is 1 to 20, n is 1 to 8, o is 5 to 40, p is 2 to 15, and x is 0 to 30. In one embodiment, the binder is $Al_{13}O_4(OH)_{24}Cl_7 \cdot 12(H_2O)$ as is described in G.M. Wolterman, et al., *Stud. Surf. Sci. and Catal.*, 76, pages 105-144 (1993), which is herein incorporated by reference. In another embodiment, one or more binders are combined with one or more other non-limiting examples of alumina materials such as aluminum oxyhydroxide, γ -alumina, boehmite, diasporite, and transitional aluminas such as α -alumina, β -alumina, γ -alumina, δ -alumina, ϵ -alumina, κ -alumina, and ρ -alumina, aluminum trihydroxide, such as gibbsite, bayerite, nordstrandite, doyleite, and mixtures thereof.

[0062] In another embodiment, the binders are alumina sols, predominantly comprising aluminum oxide, optionally including some silicon. In yet another embodiment, the binders are peptized alumina made by treating alumina hydrates such as pseudoboehmite, with an acid, preferably an acid that does not contain a halogen, to prepare sols or aluminum ion solutions. Non-limiting examples of commercially available colloidal alumina sols include Nalco 8676 available from Nalco Chemical Co., Naperville, Illinois, and Nyacol available from The PQ Corporation, Valley Forge, Pennsylvania.

[0063] The molecular sieve, in a preferred embodiment, is combined with one or more matrix material(s). Matrix materials are typically effective in reducing overall catalyst cost, act as thermal sinks assisting in shielding heat from the catalyst composition for example during regeneration, densifying the catalyst composition, increasing catalyst strength such as crush strength and attrition resistance, and to control the rate of conversion in a particular process.

[0064] Non-limiting examples of matrix materials include one or more of: rare earth metals, metal oxides including titania, zirconia, magnesia, thoria, beryllia, quartz, silica or sols, and mixtures thereof, for example silica-magnesia, silica-zirconia, silica-titania, silica-alumina and silica-alumina-thoria. In an

embodiment, matrix materials are natural clays such as those from the families of montmorillonite and kaolin. These natural clays include sabbentonites and those kaolins known as, for example, Dixie, McNamee, Georgia and Florida clays. Non-limiting examples of other matrix materials include: halloysite, kaolinite, dickite, nacrite, or anauxite. In one embodiment, the matrix material, preferably any of the clays, are subjected to well known modification processes such as calcination and/or acid treatment and/or chemical treatment.

[0065] In one preferred embodiment, the matrix material is a clay or a clay-type composition, preferably the clay or clay-type composition having a low iron or titania content, and most preferably the matrix material is kaolin. Kaolin has been found to form a pumpable, high solid content slurry, it has a low fresh surface area, and it packs together easily due to its platelet structure. A preferred average particle size of the matrix material, most preferably kaolin, is from about 0.1 μm to about 0.6 μm with a D90 particle size distribution of less than about 1 μm .

[0066] In another embodiment, the weight ratio of the binder to the matrix material used in the formation of the molecular sieve catalyst composition is from 0:1 to 1:15, preferably 1:15 to 1:5, more preferably 1:10 to 1:4, and most preferably 1:6 to 1:5. It has been found that a higher sieve content, lower matrix content, increases the molecular sieve catalyst composition performance, however, lower sieve content, higher matrix material, improves the attrition resistance of the composition.

[0067] In another embodiment, the formulated molecular sieve catalyst composition contains from about 1% to about 99%, more preferably from about 5% to about 90%, and most preferably from about 10% to about 80%, by weight of the molecular sieve based on the total weight of the molecular sieve catalyst composition.

[0068] In another embodiment, the weight percent of binder in or on the spray dried molecular sieve catalyst composition based on the total weight of the binder, molecular sieve, and matrix material is from about 2% by weight to about 30% by weight, preferably from about 5% by weight to about 20% by weight, and more preferably from about 7% by weight to about 15% by weight.

[0069] Once the molecular sieve catalyst composition is formed in a substantially dry or dried state, to further harden and/or activate the formed catalyst composition, a heat treatment such as calcination, at an elevated temperature is usually performed. A conventional calcination environment is air that typically includes a small amount of water vapor. Typical calcination temperatures are in the range from about 400°C to about 1,000°C, preferably from about 500°C to about 800°C, and most preferably from about 550°C to about 700°C, preferably in a calcination environment such as air, nitrogen, helium, flue gas (combustion product lean in oxygen), or any combination thereof.

III. Gas-Solids Reactor Systems

[0070] In the gas-solids reactor system of this invention, gas is flowed along with the solids. The gas can be inert or a chemical reactant.

[0071] In a preferred embodiment of the invention, the gas flowing through the gas-solids reactor system is a chemical reactant and the solids are catalyst particles capable of transforming the reactant into a product.

[0072] In one embodiment, the chemical reactant is an oxygenate and the catalyst particles are molecular sieves. Molecular sieves capable of converting the oxygenate to olefins are preferred, such systems typically being referred to as oxygenate to olefin reaction systems. Any of the above described molecular sieves can be used to convert oxygenates to olefins. Conventional zeolites and silicoaluminophosphates are preferred.

[0073] Oxygenates used in this invention include one or more organic compound(s) containing at least one oxygen atom. In the most preferred embodiment of the process of invention, the oxygenate in the feedstock is one or more alcohol(s), preferably aliphatic alcohol(s) where the aliphatic moiety of the alcohol(s) has from 1 to 20 carbon atoms, preferably from 1 to 10 carbon atoms, and most preferably from 1 to 4 carbon atoms. The alcohols useful as feedstock in the process of the invention include lower straight and branched chain aliphatic alcohols and their unsaturated counterparts. Non-limiting examples of oxygenates include methanol, ethanol, n-propanol, isopropanol, methyl ethyl ether, dimethyl ether, diethyl ether, di-isopropyl ether, formaldehyde, dimethyl carbonate, dimethyl ketone, acetic acid, and mixtures thereof. In the most preferred

embodiment, the feedstock is selected from one or more of methanol, ethanol, dimethyl ether, diethyl ether or a combination thereof, more preferably methanol and dimethyl ether, and most preferably methanol.

[0074] The feedstock, in one embodiment, contains one or more diluent(s), typically used to reduce the concentration of the feedstock, and are generally non-reactive to the feedstock or molecular sieve catalyst composition. Non-limiting examples of diluents include helium, argon, nitrogen, carbon monoxide, carbon dioxide, water, essentially non-reactive paraffins (especially alkanes such as methane, ethane, and propane), essentially non-reactive aromatic compounds, and mixtures thereof. The most preferred diluents are water and nitrogen, with water being particularly preferred.

[0075] The diluent is either added directly to a feedstock entering into a reactor or added directly into a reactor, or added with a molecular sieve catalyst composition. In one embodiment, the amount of diluent in the feedstock is in the range of from about 1 to about 99 mole percent based on the total number of moles of the feedstock and diluent, preferably from about 1 to 80 mole percent, more preferably from about 5 to about 50, most preferably from about 5 to about 25. In one embodiment, other hydrocarbons are added to a feedstock either directly or indirectly, and include olefin(s), paraffin(s), aromatic(s) (see for example U.S. Patent No. 4,677,242, addition of aromatics) or mixtures thereof, preferably propylene, butylene, pentylene, and other hydrocarbons having 4 or more carbon atoms, or mixtures thereof.

[0076] Other chemical reactants that can be used in this invention include any liquid reactant stream that will enter the reactor at least partially as a liquid, but will be further vaporized by contact with the circulating solid material. These reactants or feeds include refractory crudes with boiling points that extend over wide ranges and high concentrations of metals and coke. For example, one typical crude has a boiling point that ranges from about 240°F to about 1575°F, with more than half of the liquid volume boiling above 1000°F.

[0077] When the gas-solid system is a fluidized catalytic cracking (FCC) process, suitable feedstocks include conventional FCC feeds and higher boiling or residual feeds. Conventional feeds include gas oil or vacuum gas oil. These

compositions are hydrocarbon materials having a boiling range of anywhere from about 450°F to about 1025°F. These fractions are preferably low in coke precursors and heavy metals which can deactivate the catalyst. Heavy or residual feeds, i.e., boiling above 930°F, and which have a high metals content, can also be used.

[0078] In another embodiment, this invention can be used in the oxidation of n-butane to maleic anhydride. Preferably, such a reaction process uses a vanadium/phosphorus oxide catalyst as the solid catalyst particles. Examples of such a process are described in U.S. Patent Nos. 5,519,149; 5,021,588; 4,769,477; 4,699,985; 4,668,802; 4,442,226; 4,371,702, the descriptions of each being fully incorporated herein by reference.

[0079] In yet another embodiment, this invention can be used in the ammonia oxidation of propylene or propane to acrylonitrile. Preferably, such a reaction process uses a Mo or Bi containing catalyst or a V, Sb, or Sn containing catalyst as the solid catalyst particles. Examples of such a process are described in U.S. Patent Nos. 6,245,931; 5,834,394; 5,658,842; 6,166,241; 6,143,916; and 5,907,052, the descriptions of each being fully incorporated herein by reference.

IV. Process Conditions

[0080] In one embodiment of this invention, the gas and solid particles are flowed through the gas-solids reactor system at a weight hourly space velocity (WHSV) of from about 2 hr⁻¹ to about 5,000 hr⁻¹, preferably from about 5 hr⁻¹ to about 3,000 hr⁻¹, more preferably from about 10 hr⁻¹ to about 1,500 hr⁻¹, and most preferably from about 20 hr⁻¹ to about 1,000 hr⁻¹. In one preferred embodiment, the WHSV is greater than 25 hr⁻¹, and up to about 500 hr⁻¹. In this invention, WHSV is defined as the total weight per hour of the gas flowing between reactor walls divided by the total weight of the solids flowing between the same segment of reactor walls. The WHSV is maintained at a level sufficient to keep the catalyst composition in a fluidized state within a reactor.

[0081] In another embodiment of the invention, the gas and solid particles are flowed through the gas-solids reactor system at a gas superficial velocity (GSV) at least 1 meter per second (m/sec), preferably greater than 2 m/sec, more

preferably greater than 3 m/sec, and most preferably greater than 4 m/sec. The GSV should be sufficient to maintaining the solids in a fluidized state.

[0082] In yet another embodiment of the invention, the solids particles and gas are flowed through the gas-solids reactor at a solids to gas mass ratio of about 5:1 to about 75:1. Preferably, the solids particles and gas are flowed through the gas-solids reactor at a solids to gas mass ratio of about 8:1 to about 50:1, more preferably from about 10:1 to about 40:1.

V. Examples of Particular Embodiments

[0083] Figures 1-4 show various embodiments of the invention. In Fig. 1, a riser portion 10 of a reactor system is connected to a standpipe assembly 12 of the reactor system 12. The standpipe assembly 12 can be linked to a catalyst regenerator (not shown) or it can merely be used as a circulation line to recontact catalyst with feed. Fig. 1 shows that as circulation catalyst is circulated through the standpipe 12 to contact feed input into the riser portion 10, aeration fluid is injected at a point away from the wall of the standpipe portion 12.

[0084] Fig. 2 shows a standpipe arrangement 20 which includes multiple standpipe portions 21, 22. In this embodiment, the standpipe 20 has 2 points of injection, exemplified by nozzle points 23, 24, corresponding to points of injection R and r, respectively, from the centerline of the standpipe 20. In this embodiment, $r/R < 0.75$. Also shown are valves 25, 26, which are used to control the flow rate of solid material back to the reactor.

[0085] Fig. 3 shows a single standpipe arrangement 30, having microporous gas spargers 32. The microporous gas spargers are positioned appropriate distances apart to minimize bubble formation, and thereby increase apparent catalyst density in the standpipe 30.

[0086] Fig. 4 shows a horizontal section view of a standpipe 40. Located at substantially the same horizontal plane are injection orifices 42, 44 and internal sparger 46. In this embodiment, the sparger has multiple orifices (not shown) arranged to flow aeration fluid in the direction of flowing solid particles. Aeration fluid is injected through orifices 42, 44 radially inward and spaced 180 degrees in the azimuthal direction.

VI. Examples of Differential Pressure Profiles

Example 1

[0087] Solid particles were flowed through a standpipe having a 4.4375" slide valve to control the flow of solid particles through the standpipe. The standpipe included a single orifice for injecting aeration fluid into the standpipe. The solid particles were flowed through the standpipe at a mass flux, G_s , of 139 lb/ft²s. Differential pressure (DP) was measured at 11 locations from just above the slide valve (DP SP1) to the inlet of the standpipe (DP SP11). The catalyst had an apparent density ($\Delta P/L$) of 17 lb/ft³. Dynamic data was collected over a period of 5 minutes. The results are shown in Fig. 5.

Example 2

[0088] Solid particles were flowed through a standpipe as in Example 1, except that the standpipe had a 3.5" slide valve; and the particles were flowed at a G_s of 121 lb/ft²s. The standpipe also had two orifices spaced about 180 degrees from one another at approximately the same elevation. The catalyst had an apparent density ($\Delta P/L$) of 22 lb/ft³. Dynamic data was collected over a period of 5 minutes. The results are shown in Fig. 6.

Example 3

[0089] Solid particles were flowed through a standpipe as in Example 1, except that the particles were flowed at a G_s of 121 lb/ft²s, and the standpipe had three orifices spaced about 120 degrees from one another at approximately the same elevation. The catalyst had an apparent density ($\Delta P/L$) of 23 lb/ft³. Dynamic data was collected over a period of 5 minutes. The results are shown in Fig. 7.

Example 4

[0090] Solid particles were flowed through a standpipe as in Example 1, except that the particles were flowed at a G_s of 121 lb/ft²s. This standpipe further included the use of a sparger, and was similar to the embodiment shown in Fig. 4. The catalyst had an apparent density ($\Delta P/L$) of 23 lb/ft³. Dynamic data was collected over a period of 5 minutes. The results are shown in Fig. 8.

[0091] The data indicates that the use of at least two orifices substantially reduces differential pressure fluctuations compared to a single orifice. In general, additional orifices further reduce the differential pressure fluctuations, with the addition of a sparger arrangement being more preferred. Moreover, the 21 lb/ft³ density of solid particles in the standpipe of was significantly greater than the 17 lb/ft³ cited in Example 1, where only a single aeration orifice was used.

[0092] Having now fully described this invention, it will be appreciated by those skilled in the art that the invention can be performed within a wide range of parameters within what is claimed, without departing from the spirit and scope of the invention.

Claims:

1. A method of aerating solid particles in a standpipe of a gas-solids reactor system, comprising:
 - a) providing a standpipe having an internal wall and solid particles within the standpipe, and
 - b) injecting aeration fluid at a point away from the internal wall, wherein at least 25% of the aeration fluid is injected at a point away from the internal wall so as to aerate the solid particles in the standpipe.
2. The method of claim 1, wherein at least 50% of the aeration fluid is injected at a point away from the internal wall.
3. The method of claim 2, wherein at least 75% of the aeration fluid is injected at a point away from the internal wall.
4. The method of any of the preceding claims, wherein the standpipe has a centerline and the aeration fluid is injected into the standpipe at a point such that at least a portion of the fluid is injected at a location $r/R < 0.75$, with r being defined as the radial distance from the centerline of the standpipe to the point of aeration fluid injection, and R being defined as the radial distance from the centerline of the standpipe to the internal wall of the standpipe.
5. The method of claim 4, wherein $r/R < 0.6$.
6. The method of claim 5, wherein $r/R < 0.5$.
7. The method of the preceding claims, wherein the solid particles in the standpipe have an apparent density of at least 50% of the maximum apparent density of the solid particles.

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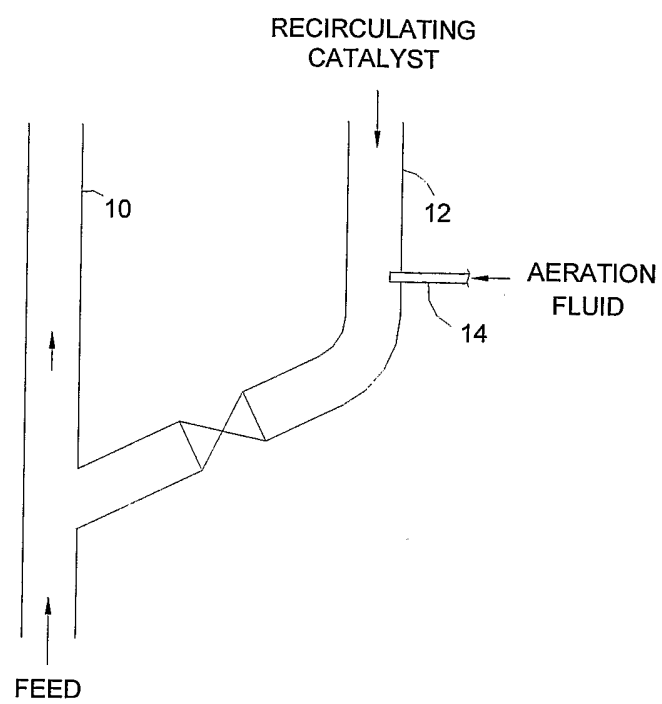
8. The method of claim 7, wherein the solid particles in the standpipe have an apparent density of at least 60% of the maximum apparent density of the solid particles.
9. The method of claim 8, wherein the solid particles in the standpipe have an apparent density of at least 80% of the maximum apparent density of the solid particles.
10. The method of any of the preceding claims, wherein the solid particles flow through the standpipe at a velocity of from 0.25 m/sec to 5 m/sec.
11. The method of claim 10, wherein the solid particles flow through the standpipe at a velocity of from 0.75 m/sec to 4 m/sec.
12. The method of claim 11, wherein the solid particle flow through the standpipe at a velocity of from 1 m/sec to 3 m/sec.
13. The method of the preceding claims, wherein the solid particles flow through a reactor portion of the gas-solids reactor system at a velocity of from 1 m/sec to 30 m/sec.
14. The method of claim 13, wherein the solid particles flow through the reactor portion of the gas-solids reactor system at a velocity of from 3 m/sec to 25 m/sec.
15. The method of claim 14, wherein the solid particles flow through the reactor portion of the gas-solids reactor system at a velocity of from 5 m/sec to 20 m/sec.
16. The method of any of the preceding claims, wherein the standpipe is in an oxygenate to olefin reactor system.

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17. The method of any of claims 1 to 15, wherein the standpipe is in a fluidized catalytic cracking reactor system.
18. The method of any of claims 1 to 15, wherein the standpipe is in a reactor system for oxidation of n-butane to maleic anhydride.
19. The method of any of claims 1 to 15, wherein the standpipe is in a reactor system for ammonia oxidation of propylene or propane to acrylonitrile.
20. The method of any of the preceding claims, wherein the standpipe has an internal diameter of at least 2 feet.
21. The method of any of the preceding claims, wherein the solid particles in the standpipe have an apparent density of at least 20 lb/ft³.
22. The method of any of the preceding claims, wherein aeration fluid is further injected through at least two aeration orifices located at different azimuthal positions on the standpipe.
23. The method of claim 22, wherein the aeration orifices are separated at a distance of from $\pi/5$ to π radians.
24. The method of claim 23, wherein the aeration orifices are separated at a distance of from $\pi/4$ to $2\pi/3$ radians.
25. The method of any of claims 22 to 24, wherein the aeration fluid is injected through at least one aeration orifice in an azimuthal direction around a circumference of the internal wall.
26. The method of any of the preceding claims, wherein aeration fluid is further injected into the standpipe in inwardly radial directions of the

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internal wall, through at least two aerations orifices located within 1/2 foot of a horizontal baseline through the standpipe.

**Fig. 1**

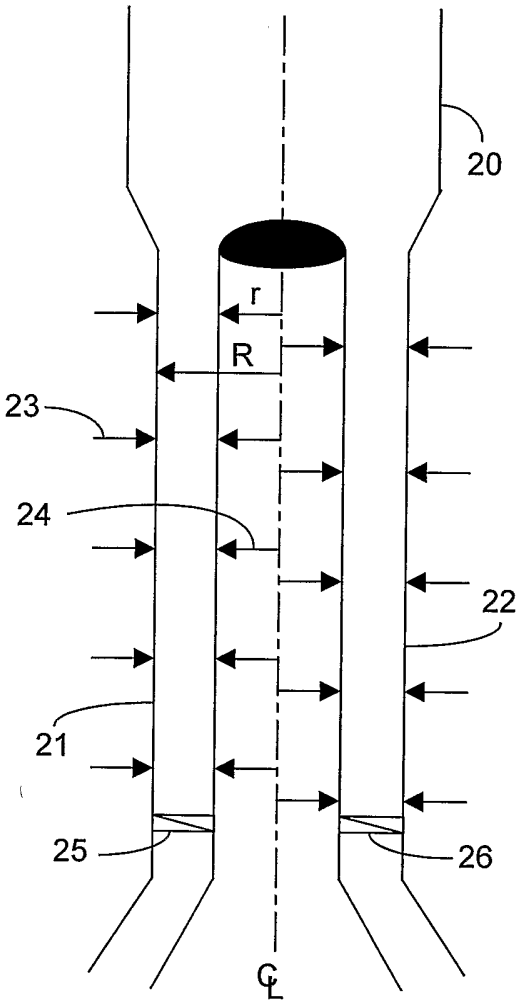


FIG. 2

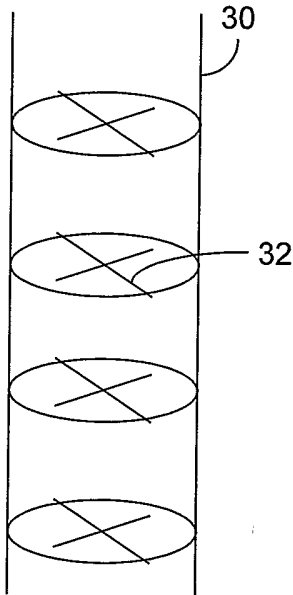
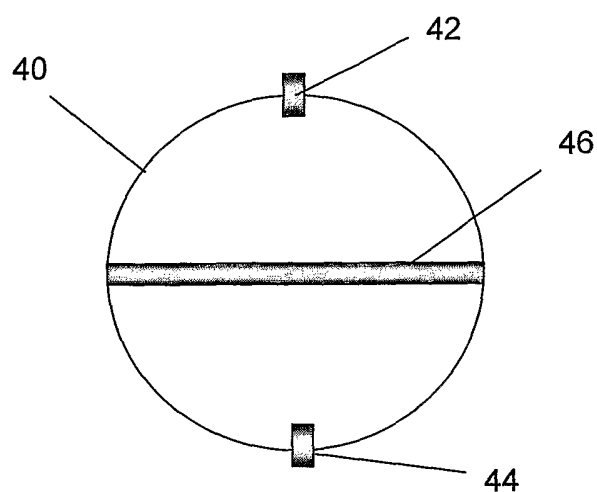
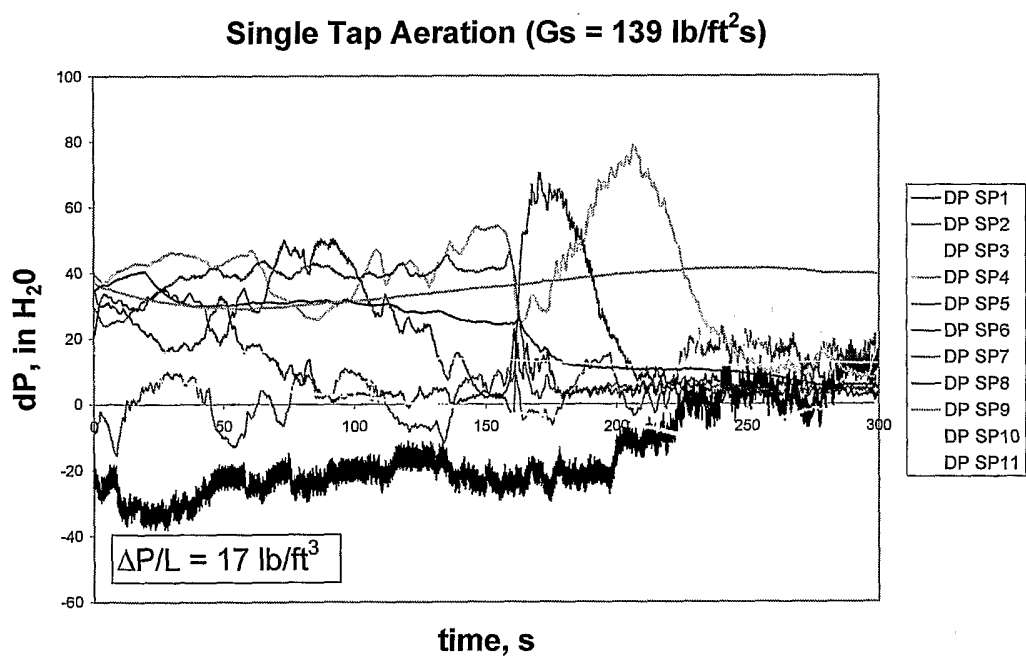
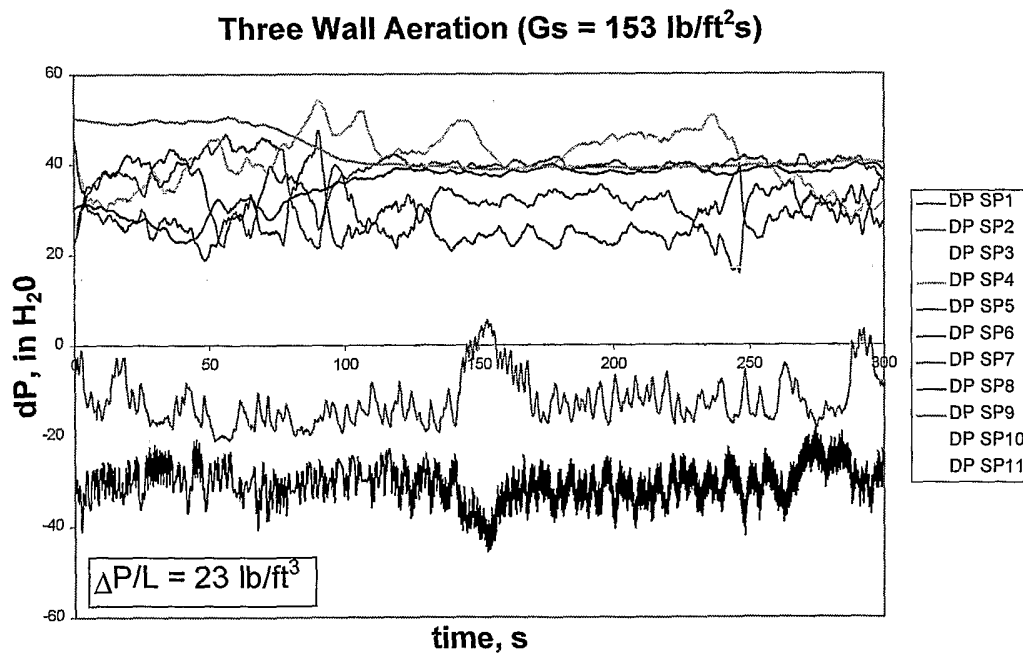
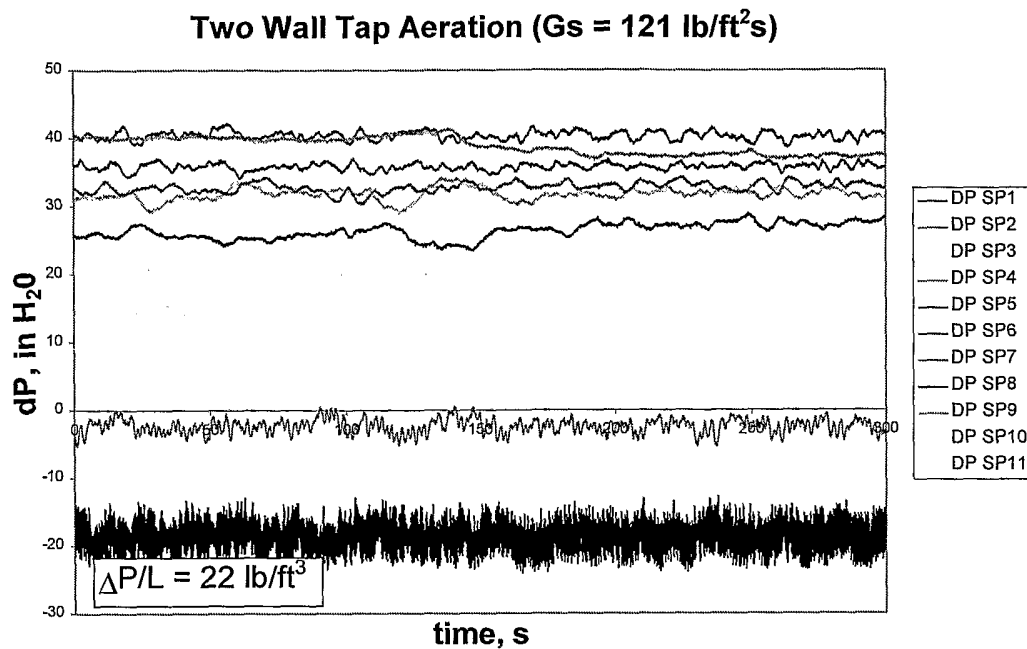
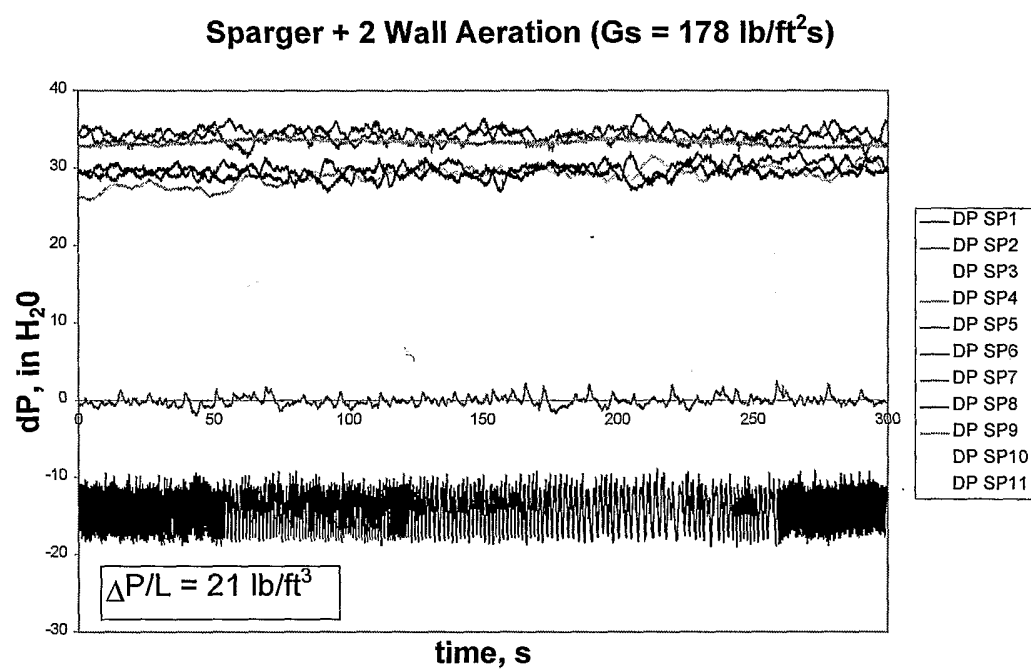


FIG. 3

**FIG. 4****FIG. 5**



**FIG. 8**