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(54) IMPROVEMENTS RELATING TO PYROLYSIS PROCESSES UTILIZING A PARTICULATE HEAT SOURCE

(71) We, OCCIDENTAL PETROLEUM CORPORATION, a Corporation organised and existing under the Laws of the State of California, United States of America of 10889 Wilshire Boulevard, Los Angeles, California 90024, United States of America do hereby declare the invention, for which we pray that a patent may be granted to us and the method by which it is to be performed, to be particularly described in and by the following statement:

The increasing scarcity of fluid fossil fuels such as oil and natural gas is causing much attention to be directed towards converting solid carbonaceous materials such as coal, oil shale, tar sands, uintaite and solid waste to liquid and gaseous hydrocarbons by pyrolysis. Pyrolysis can occur under nonoxidizing conditions in a pyrolysis reactor in the presence of a particulate source of heat to yield as products pyrolytic vapors containing hydrocarbons and a particulate carbon-containing solid residue. The particulate source of heat for effecting the pyrolysis of the carbonaceous material can be obtained by oxidizing carbon in the particulate carbon-containing solid residue in a combustion chamber.

There are many problems associated with this use of a pyrolysis reactor and a combustion chamber in combination for obtaining hydrocarbons from solid carbonaceous materials. One of these problems is caking of coal along the walls of the pyrolysis reactor when the carbonaceous material is an agglomerative coal, particularly an Eastern United States coal, which experience shows to have a tendency to agglomerate in a reactor, especially along the walls of the reactor.

Another problem concerns transferring the particulate carbon-containing solid product from the pyrolysis reactor to the combustion chamber while at the same time preventing oxygen that is present in the combustion chamber from entering the pyrolysis reactor. If oxygen manages to leak into the pyrolysis reactor, the value of the hydrocarbon product is reduced and moreover, a violent explosion may occur.

A third problem concerns the need to maximize production of carbon dioxide and minimize production of carbon monoxide in the combustion zone in order to maximize recovery of the heating value of the carbon-containing solid residue during oxidation. The kinetics and thermodynamic equilibria of the oxidation of carbon favour increased production of carbon monoxide relative to carbon dioxide at temperatures greater than about 1200°F. (650°C.) at long residence times when there is a stoichiometric deficiency of oxygen. Because pyrolysis of carbonaceous materials often is conducted at temperatures greater than 1200°F. (650°C.) and can approach temperatures higher than 2000°F. (1100°C.), it is necessary to form a particulate source of heat having temperatures greater and often considerably greater than 1200°F. (650°C.). Moreover, the particulate carbon-containing solid residue is only partly oxidized in a stoichiometric deficiency of oxygen to form the particulate source of heat. Thus production of carbon monoxide inevitably occurs during the oxidation of the particulate carbon-containing solid residue. The carbon monoxide formed represents a loss of thermal efficiency of the process.

Therefore, there is a need for a process and an apparatus for obtaining values from a solid carbonaceous material by pyrolysis which are useful for agglomerative coals; which, when a particulate carbon-containing solid residue of pyrolysis of the carbonaceous material is oxidized to form a particulate source of heat to pyrolyze the carbonaceous material, prevent oxygen from entering into the pyrolysis reaction; and which maximize production of carbon dioxide while minimizing production of carbon monoxide.

In one aspect the invention provides a process for pyrolysis of solid carbonaceous materials by heat transferred thereto by a particulate source of heat to yield a particulate carbon-containing solid residue as a product of pyrolysis, the particulate source of heat being formed by oxidizing at least a portion of the particulate carbon-containing solid residue, characterised in that the particulate source of heat is generated by transporting at least a portion of the particulate carbon-containing solid residue formed by pyrolysis of the solid carbonaceous material to a fluidized bed around a substantially vertically oriented, open conduit in open communication with a substantially vertically oriented riser, the conduit and riser comprising a first combustion zone; educting solid residue from the fluidized bed into the first combustion zone and transporting it therethrough to a second combustion zone by injecting a transport gas upwardly into the conduit, while introducing a gaseous source of oxygen into said combustion zones to oxidize carbon in the solid residue.

In this process the fluidized bed of particulate carbon-containing solid residue of pyrolysis that is disposed around the conduit that with the riser, comprises the first combustion zone, serves as a barrier to backflow of the oxygen source from the first combustion zone, while by effecting oxidation of the carbon-containing solid residue in two stages in the respective combustion zones carbon monoxide produced in the first combustion zone can be at least partly converted to carbon dioxide in the second combustion zone to enhance the thermal efficiency of the generation of the particulate heat source. In preferred practice, the amount of oxygen introduced into the second combustion zone is equal to at least 50% of the molar amount of carbon monoxide produced in the first combustion zone, to maximise conversion of the carbon monoxide to carbon dioxide.

Preferably the said conduit is spaced apart from said riser and the particulate carbon-containing solid residue is fluidized in the fluidized bed by an upward flow of a fluidizing gas that passes into the riser through the space between the riser and the conduit. The fluidizing gas desirably contains oxygen.

The formed particulate source of heat may be separated from the gaseous combustion products of the second combustion zone in a cyclone separation zone. Alternatively, the second combustion zone may comprise a cyclone oxidation-separation zone.

In preferred embodiments of the invention the pyrolysis process is a so-called flash pyrolysis step, performed by continuously transporting particulate solid carbonaceous material feed contained in a carrier gas which is substantially non-deleteriously reactive with respect to products of pyrolysis to a vertically oriented, descending flow pyrolysis reactor containing a pyrolysis zone operated at a temperature below about 2000°F. (1100°C.); feeding the particulate source of heat at a temperature above the pyrolysis temperature to the pyrolysis reactor at a rate sufficient to maintain said pyrolysis zone at the pyrolysis temperature; forming a turbulent mixture of the particulate source of heat, particulate solid carbonaceous material feed and the carrier gas to pyrolyze the solid carbonaceous material feed and yield a pyrolysis product stream containing as solids, the particulate source of heat and a particulate carbon-containing solid residue of pyrolysis, and a vapour mixture of carrier gas pyrolytic vapours comprising hydrocarbons; and passing the pyrolysis product stream from the pyrolysis reactor to a first separation zone to separate solids from the vapour mixture.

The first separation zone may be a cyclone separation zone.

The process is preferably performed with the use of a pyrolysis reactor having a solids feed inlet for the solid carbonaceous material feed and a vertically oriented chamber surrounding the upper portion of the pyrolysis reactor, the inner peripheral wall of said chamber forming an overflow weir to a vertically oriented mixing zone of the reactor, the particulate solid carbonaceous material being transported in a carrier gas to the solids feed inlet and thence being injected into the mixing zone, the particulate source of heat being fed to the said vertically oriented chamber and being fluidized in such chamber by a flow of a fluidizing gas substantially nondeleteriously reactive with respect to the products of pyrolysis, the fluidized particulate source of heat discharging over said weir and downwardly into said mixing zone to form a turbulent mixture with the particulate solid carbonaceous material, said mixture passing downwardly from the mixing zone to the pyrolysis zone of the pyrolysis reactor to pyrolyze the solid carbonaceous material.

The invention also provides an apparatus for forming a particulate solid source of heat from a particulate carbon-containing solid residue of pyrolysis of a carbonaceous material for pyrolysis of the solid carbonaceous material, comprising a vessel for containing a fluidized bed of said particulate carbon-containing solid residue of pyrolysis around an open, substantially vertically oriented conduit; a substantially vertically oriented riser in open communication with the conduit, said conduit and said riser comprising a first combustion chamber; a second combustion chamber in communication with the riser; means for introducing particulate carbon-containing solid residue of pyrolysis into the fluidized bed; means for injecting a transport gas upwardly into the conduit to educt carbon-containing residue from the fluidized bed upwardly into the conduit; and means for introducing oxygen into the combustion chambers to oxidise carbon in the said solid residue to form the particulate source of heat.

Such apparatus is particularly useful for combination with a pyrolysis reactor utilising a particulate solid source of heat. Accordingly, in another aspect the invention provides an apparatus for pyrolysis of solid carbonaceous materials, comprising a descending flow pyrolysis reactor; means for forming a turbulent mixture of a particulate source of heat and a solid carbonaceous material contained in a carrier gas for introduction into the pyrolysis reactor to pyrolyze the solid carbonaceous feed to form a pyrolysis product stream containing a vapour mixture and, as solids, the particulate source of heat and a particulate carbon-containing solid residue of pyrolysis; means for transferring the pyrolysis product stream from the pyrolysis reactor to a first separator adapted for separating solids from the vapour mixture in the pyrolysis product stream; means for passing the separated solids from the first separator to a vessel for containing a fluidized bed for the separated solids around an open, substantially vertically oriented conduit, a substantially vertically oriented riser being in open communication with the conduit and comprising therewith a first combustion chamber; a second combustion chamber in communication with the riser; means for injecting a transport gas upwardly into the conduit to educt solids from the fluidized bed upwardly into the conduit, to transport the solids through the first combustion chamber to the second combustion chamber; and means for introducing oxygen into the combustion chambers to oxidise carbon in said solids thereby to heat the solids to form the particulate source of heat and to produce gaseous combustion products; means for transferring the particulate source of heat from the second combustion chamber to a second separator adapted for separating the particulate source of heat from the gaseous combustion products; and means for transferring the separated particulate source of heat from the second separator to the pyrolysis reactor.

Preferably the said conduit is spaced apart from said riser. The first and second separators may conveniently both be cyclone separators although in a modified arrangement the second combustion chamber and the second separator are integrated and constituted by a cyclone oxidation-separation device.

In a preferred embodiment of the apparatus, the pyrolysis reactor contains a substantially vertically oriented mixing section and a substantially vertically oriented pyrolysis section, and has a solids feed inlet, a substantially vertically oriented chamber surrounding the upper portion of the reactor and having an inner peripheral wall that forms an overflow weir to the vertically oriented mixing section, and the means for forming a turbulent mixture comprises means for feeding particulate source of heat to the vertically oriented chamber; means for introducing a fluidizing gas into the chamber to maintain the particulate source of heat therein in a fluidized state; and means for injecting the solid carbonaceous feed contained in the carrier gas from the solids feed inlet into the mixing section to form the resultant turbulent mixture.

In another aspect the invention provides an apparatus for pyrolysis of agglomerative coals, comprising a descending flow pyrolysis reactor containing a substantially vertically oriented mixing section, a substantially vertically oriented pyrolysis section, a solids feed inlet, and a substantially vertically oriented chamber surrounding the upper portion of the reactor, the inner peripheral wall of the chamber forming an overflow weir to the mixing section, wherein an agglomerative coal feed contained in a carrier gas is combined with a particulate source of heat under turbulent flow conditions in the pyrolysis section of the pyrolysis reactor to yield a pyrolysis product stream containing, as solids, the particulate source of heat and a particulate carbon-containing solid residue of pyrolysis, and a vapour mixture; means for feeding the particulate source of heat to the vertically oriented chamber; means for introducing a fluidizing gas into the chamber to maintain the particulate source of heat therein in a fluidized state; means for passing a coal feed from the solids feed inlet into the mixing section; a first cyclone separator in communication with the pyrolysis reactor for separating solids in the pyrolysis product stream from the vapour mixture in that stream; a vessel for containing a fluidized bed of the separated solids around an open, substantially vertically oriented conduit, a substantially vertically oriented riser being in open communication with the vertically oriented conduit and separated therefrom, said conduit and riser comprising a first combustion chamber; a second combustion chamber in communication with the riser; means for injecting a transport gas upwardly into the conduit to educt solids from the fluidized bed upwardly into the conduit and to transport said solids through the first combustion chamber to the second combustion chamber; means for introducing oxygen into said combustion chambers to oxidize carbon in the solids to heat the solids and to form the particulate source of heat with attendant formation of combustion products; a dipleg from the first cyclone separator to the fluidized bed for transferring the separated solids from the first cyclone separator to the fluidized bed; a second cyclone separator in communication with the second combustion chamber for separating the particulate source of heat from the gaseous combustion products; and the dipleg from the second cyclone separator to the chamber surrounding the upper portion of the pyrolysis reactor for transferring the particulate source of heat to the pyrolysis reactor.

Further preferable features of the process of the invention and of the apparatus thereof will be discussed and described in the following, with reference to the accompanying drawings in which:

Figure 1 illustrates diagrammatically a process and an apparatus embodying features of this invention; and

Figure 2 is an enlarged and more detailed view of the region marked 2 in Figure 1.

The drawings illustrate a pyrolysis unit 8 comprising a descending flow pyrolysis reactor 10 which has a substantially vertically oriented mixing section or zone 12 and a substantially vertically oriented pyrolysis section or zone 14 below the mixing section. Arrow 16 shows the approximate extent of the pyrolysis section. The reactor has an elbow 18 towards the end of the pyrolysis section, by which it can be supported. The lower end 20 of the reactor terminates in a separation zone such as first cyclone separator 22.

A generally upright annular solids feed inlet 24 terminating within the mixing section 12 and constricted at its end to form a nozzle 26 is provided for introducing a solid carbonaceous material into the mixing region.

The upper end 28 of the reactor is open and of larger diameter than the nozzle 26, thereby leaving an annular gap 30 between the upper end 28 of the reactor and the nozzle 26. A vertically oriented fluidising chamber or well 32 surrounds the upper portion of the reactor and is formed by a preferably annular section 34 which connects the wall 36 of the solids feed inlet above where the wall constricts to form the nozzle 26 and the upper portion 28 of the reactor. The chamber 32 surrounds the nozzle 26 and a portion of the upper wall 28 of the reactor. The inner peripheral wall of the chamber 32 is formed by the upper wall 28 of the reactor and serves as an overflow weir to the mixing section 12 of the reactor 10.

A second vertically oriented solids inlet 38 terminates in the annular fluidizing chamber 32, preferably at a level below the top edge 40 of the pyrolysis reactor 10.

There is a gas inlet 42 to the bottom of the fluidizing chamber for a fluidizing gas. Means are provided such as a cylindrical, horizontally oriented, perforated plate 44 positioned towards the bottom of the fluidizing chamber below the end of the second inlet for distributing the fluidizing gas so that the fluidizing gas flows upwardly through the fluidizing chamber.

The first cyclone separator 22 serves to separate a particulate carbon-containing solid residue of pyrolysis from the gaseous products of pyrolysis.

The particulate source of heat for the pyrolysis reactor is formed by oxidizing at least a portion of the particulate carbon-containing solid residue in a combustion unit 50. The combustion unit includes a vessel 52 that in operation contains a fluidized bed 60 of the particulate carbon-containing solid residue of pyrolysis around an open, substantially vertically oriented conduit or tube 54. There is a gas inlet 56 for a transport gas at the base of the vessel 52, which inlet narrows down to form a vertically oriented nozzle 58 for injection of the transport gas directly upwardly into the open conduit 54. The fluidized bed 60 of carbon-containing solid residue is fluidized by a fluidizing gas entering the vessel 52 through a gas inlet 62 at the base of the vessel. The fluidizing gas is distributed throughout the fluidized bed by means of a second, horizontally oriented perforated distributor plate 64.

The top 66 of the vessel 52 tapers upwardly and inwardly to connect to a vertically oriented riser 68. The riser and conduit comprise a first combustion zone or chamber. The riser couples the vessel 52 to a second combustion zone or chamber 70. The conduit 54 is below the riser 68 and the top edge 72 of the conduit is spaced apart from the riser so that an annular gap of space 74 is formed between the inlet 76 to the riser and the top edge 72 of the conduit. The top portion 71 of the conduit can be tapered inwardly so that the diameter of the conduit at its top edge is smaller than the diameter of the riser.

A vertically oriented standpipe or dipleg 78 having stripping gas inlets 120 extends from the bottom of the first cyclone separator 22 into the vessel 52 below the top 80 of the fluidized bed of carbon-containing solid residue. Solids separated by the first cyclone separator are transferred through this dipleg into the vessel.

There is an inlet 82 at the upper portion of the riser 68 for introduction of a source of oxygen into the second combustion chamber 70. The second combustion chamber is in open communication with a second separator such as cyclone separator 84. This separator serves to separate a particulate source of heat generated in the combustion unit 50 from any combustion gases present in the combustion unit. The particulate source of heat is transferred from the second cyclone separator 84 to the second inlet 38 of the pyrolysis reactor through a vertically oriented dipleg or standpipe 86 originating at the bottom of the second cyclone separator 84 and terminating in the second inlet 38. The length of the standpipe 86 is chosen to balance the accumulation of differential pressures throughout the remainder of the system. Inlets 88 for a stripping or aeration gas are provided along the length of the standpipe 86.

In summary, what has been described is an apparatus for pyrolysis of a solid carbonaceous material comprising two main units, a pyrolysis unit 8 and a combustion unit 50. These two units are coupled by two cyclone separators 22, 84 and two vertically oriented standpipes or diplegs 78, 86 which allow carbon-containing solid residue to be transferred from the pyrolysis unit to the combustion unit and particulate source of heat to be transferred from the cumbustion unit to the pyrolysis unit, respectively.

In the process of this invention as exemplified by this embodiment, a particulate solid carbonaceous material is subjected to flash pyrolysis by transporting the particulate solid carbonaceous material feed contained in a carrier gas through the first feed inlet 24 to the feed nozzle 26 and thus to the pyrolysis reactor 10. The carrier gas is substantially non-deleteriously reactive with respect to the products of pyrolysis and may serve as a diluent to prevent self-agglomeration of the carbonaceous material.

As used herein, by a "nondeleteriously reactive" gas there is meant a gas stream which is substantially free of free oxygen. Although the gas may contain constituents that react under nonoxidizing conditions with pyrolysis products to upgrade their value, the gas should not contain constituents that degrade pyrolysis products. The carrier gas may, for instance, be the off-gas product of pyrolysis; steam, which will react under suitable conditions with char or coke formed from pyrolysis to yield, by water-gas shift reactions, hydrogen that serves to react with and stabilize unsaturates in the products of pyrolysis; and desired inert gas; or mixtures thereof. The carrier gas can, for instance, be synthesis gas, especially a hydrogen-enriched synthesis gas.

The carbonaceous material may be treated before it is fed to the pyrolysis reactor by processes such as removal of inorganic fractions by magnetic separation and classification, particularly in the case of municipal solid waste. The carbonaceous material also can be dried to reduce its moisture content. The solid carbonaceous material is usually comminuted to increase the surface area available for pyrolysis.

Preferably a substantial portion of the carbonaceous material is of a particle size of less than 1000 microns to present a large surface-to-volume ratio to obtain rapid heating of the material in the pyrolysis zone. Rapid heating results in improved yields of hydrocarbons. When the carbonaceous material is an agglomerative coal, the particle size is preferably mainly less than 250 microns because agglomerative coals are well known to plasticize and agglutinate at relatively low temperatures i.e., 400 to 850°F. (200 - 450°C.). An agglomerative coal should therefore be rapidly heated through the plastic state before it strikes the wall of a pyrolysis reactor to prevent caking on the reactor walls. Because the rate at which a coal particle can be heated increases as particle size decreases, it is important that an agglomerative coal be comminuted to 250 microns or less, depending on the size and configuration of the pyrolysis reactor, so that substantially all the coal particles have passed through the plastic state and have become non-tacky by the time the coal particles strike a reactor wall. For example, when a bituminous high-volatile C coal which agglomerates at temperatures above 500°F. (260°C.) is pyrolyzed at a temperature of 1075°F. (580°C.) in a 10 inch (250 mm) diameter pyrolysis reactor of the design shown in Figure 1 and described below, the coal should be comminuted to a size less than 250 microns in diameter to prevent caking on the reactor walls. Coal particles larger than 250 microns in diameter could strike the reactor walls before passing through the plastic state.

The carbonaceous material introduced into the pyrolysis reactor is preferably substantially free of fines less than 10 microns in diameter, because carbon-containing solid residue fines resulting from pyrolysis of the carbonaceous material have a tendency to be carried into and contaminate the liquid hydrocarbon products.

Simultaneously with the introduction of the carbonaceous material feed, there is introduced a particulate source of heat into the fluidizing chamber 32 through the second vertically oriented inlet 38. Because in the preferred embodiment the second inlet 38 terminates below the top edge 40 of the pyrolysis reactor 10, incoming particulate source of heat builds up in the fluidizing chamber below the weir 28 to form a solids seal. The particulate source of heat in chamber 32 is maintained in a fluidized state in the chamber by introduction of a fluidizing gas stream through the gas inlet 42. The fluidizing gas is distributed by the distributor plate 44 to maintain the particulate source of heat in a fluidized state throughout the chamber. As additional particulate source of heat is introduced into the chamber the particulate source of heat passes over the upper end 40 of the weir and through the opening 30 between the weir and the nozzle 26, into the mixing zone 12 of the pyrolysis reactor 10 with aid of fluidizing gas. An advantage of this weir-like configuration is that a substantially steady flow of fluidized particulate source of heat enters the mixing section because the mass of the particulate source of heat backed up behind the weir of the reactor damps minor fluctuations in the flow of the particulate source of heat.

In the mixing zone of the pyrolysis reactor, the carbonaceous material contained in the carrier gas is discharged from the nozzle as a fluid jet 112 expanding towards the reactor wall at an angle of about 20° or less as shown by dotted lines 89 which represent the periphery of the fluid jet. Once the particulate source of heat is inside the mixing section, it falls into the path of the fluid jet 112 of the carbonaceous material feed stream and carrier gas coming from the nozzle and is entrained thereby, yielding a resultant turbulent mixture of the particulate source of heat, particulate solid carbonaceous material feed, and the carrier gas. The jet has a free core region 113 of carbonaceous material, as delineated by the V-shaped dotted line 114, extending considerably into the reactor, but as the jet expands, the particulate source of heat present is entrained with mixing of the carbonaceous material in the portion of the fluid jet 112 around the free core region 113. The particulate source of heat along the periphery 89 of the fluid jet preferably heats the carbonaceous material in the case of an agglomerative coal to a temperature above the temperature at which the coal is tacky. In the region between the reactor walls and the fluid jet 112, there is unentrained particulate source of heat.

This mixing of the particulate source of heat with the solid carbonaceous material in the mixing zone 12 initiates heat transfer from the particulate solid source of heat to the carbonaceous material, causing pyrolysis in the pyrolysis section 14 of the pyrolysis reactor 10. Pyrolysis is a combination of vaporization and cracking reactions. As the vaporization and cracking ractions occur, condensible and noncondensible hydrocarbons are generated from the carbonaceous material with an attendant production of a carbon-containing solid residue such as coke or char. An effective pyrolysis time is less than about 5 seconds, and preferably ranges from 0.1 to 3 seconds, to maximize yield of middle distillates. Middle distillates are the middle boiling hydrocarbons, i.e., C_5 hydrocarbons to hydrocarbons having a boiling end point of about 950°F. (510°C.). These hydrocarbons are useful for the production of gasoline, diesel fuel, heating fuel, and the like.

As used herein, "pyrolysis time" means the time from when the carbonaceous material contacts the particulate source of heat until the pyrolytic vapors produced by pyrolysis are separated from the particulate source of heat in the first separation zone 22, as described below.

A convenient measure of pyrolysis time is the average residence time of the carrier gas in the pyrolysis section 14 of the pyrolysis reactor and the first separator 22. Sufficient pyrolysis time must be provided to heat the carbonaceous material to the pyrolysis temperature.

An advantage of the pyrolysis reactor shown in the drawings is that the turbulent flow causes the solid carbonaceous material feed to be heated rapidly, which improves yields. In the case of agglomerative coals, buildups of coal particles on the reactor walls are prevented by the rapid heating and turbulent flow. Preferably the particulate source of heat enters the mixing section 12 at a rate of flow less than turbulent and the solid carbonaceous material enters the mixing section through the nozzle under turbulent flow conditions at a rate sufficiently high for the resultant mixed stream from these two inlet streams to be turbulent. Turbulent flow results in intimate contact between the solid carbonaceous material and the particulate source of heat particles, thereby yielding rapid heating of the carbonaceous material. In the case of an agglomerative coal, the turbulence results in mixing of the particulate source of heat with the coal particles in the inner portion of the fluid jet, thereby quickly heating these coal particles through the tacky/plastic state. As used herein, "turbulent" means that the stream has a Reynolds flow index Number greater than 2000 as calculated by the velocity of the carrier gas at operating conditions. Laminar flow in the pyrolysis reactor tends to severely limit the rate of heat transfer within the pyrolysis zone. Process parameters such as the nozzle diameter and mass flow rate of the carbonaceous material and its carrier gas are varied to maintain the flow rate of the particulate stream entering the first inlet in the turbulent region.

The end of the solids feed inlet is preferably cooled, e.g. by water, when pyrolyzing an agglomerative coal because otherwise the inlet might be heated above the point at which the coal becomes tacky, by heat transfer from the particulate source of heat surrounding the end of the solids feed inlet.

Although the drawings show a solids feed inlet 24 having a nozzle 26 at its end to achieve high inlet velocities into the mixing region, a nozzle type inlet is not required alternatively, the carbonaceous material and its carrier gas can be supplied at a sufficient velocity to the inlet 24 so that the resultant mixture is under turbulent flow without need for a nozzle.

The hot particulate solid source of heat is supplied at a rate and a temperature consonant with maintaining a temperature in the pyrolysis zone suitable for pyrolysis. Pyrolysis initiates at about 600°F. (315°C.) and may be carried out at temperatures above 2000°F. (1100°C.). Preferably, however, pyrolysis is conducted at a temperature ranging from 900°F. (480°C.) to 1400°F. (760°C.) to maximize the yield of middle boiling point hydrocarbons. Higher temperatures, by contrast, enhance gasification reactions. The maximum temperature in the pyrolysis reactor is limited by the temperature at which the inorganic portion of the particulate source of heat or carbonaceous material softens with resultant fusion or slag formation.

Depending upon pyrolysis temperature, the weight ratio of particulate solid source of heat to carbonaceous material is preferably in the range 2 to 20 at the entry to the reactor. At ratios within this range, the particulate source of heat is introduced to the reactor at a temperature ranging from about 100 to about 500°F. (55 - 280°C.) above the desired pyrolysis temperature.

For economy the amount of fluidizing gas injected through inlet 42 into the fluidizing chamber is maintained at as low a level as possible subject to the constraint that the particulate source of heat be maintained in a fluidized state. Preferably at least a portion of the fluidizing gas is admitted into the mixing section of the reactor to prevent eddy formations with resultant back-mixing of partly spent particulate source of heat. The quantity of carrier gas injected with the solid carbonaceous material is that which maintains turbulent flow during the progress of the solid carbonaceous material through the plastic state in the case of an agglomerative coal. Sufficient carrier gas must be injected to prevent undesirable pressure fluctuations due to flow instabilities. The amount of gas employed to transport the solid carbonaceous material is sufficient to avoid plugging in the reactor, and normally in excess of that amount to dilute the solid materials and prevent self agglomeration in the case of an agglomerative coal.

Generally high solids content in the pyrolysis feed stream is desired to minimize equipment size and cost. However, preferably the resultant turbulent mixture contains sufficient carrier gas for the mixture to have a solids content ranging from 0.1 to 10% by volume based on the total volume of the stream, to provide turbulence for rapid heating of the carbonaceous material and to dilute the carbonaceous material and help prevent self-agglomeration, particularly when processing an agglomerative coal. Rapid heating results in high yields and prevents agglutination of agglomerative coals.

The size and configuration of the pyrolysis reactor are chosen to maintain the desired residence time for the pyrolysis reaction. Generally, as the pyrolysis temperature is reduced, longer residence times are used to maintain the desired yield of volatilized hydrocarbons.

For economy, the pressure in the pyrolysis reactor is typically greater than atmospheric to compress the vapors formed during pyrolysis so that low volume separation equipment downstream of the reactor can be used.

A pyrolysis product stream is passed from the end 20 of the pyrolysis reactor 10 to the first cyclone separator 22. The pyrolysis product stream contains, as solids, the particulate source of heat and the particulate carbon-containing solid residue of pyrolysis, and a vapor mixture of carrier gas and pyrolytic vapors comprising noncondensible hydrocarbons and condensible hydrocarbons. Preferably the first cyclone separator is in open communication with the lower end 20 of the pyrolysis reactor so that a quick separation of the vapors from the solids can be effected to minimize pyrolysis time and so that the vapors can be quenched to prevent cracking reactions from occurring which tend to decrease the recovery of middle distillates from the pyrolytic vapor. In the cyclone separator 22 at least the bulk of the solids are separated from the vapor mixture. The vapor mixture contains pyrolytic vapors containing volatilized hydrocarbons, inert carrier gases, and nonhydrocarbon components such as hydrogen sulfide which may be generated in the pyrolysis reaction.

The volatilized hydrocarbons produced by pyrolysis consist of condensible hydrocarbons which may be recovered by contacting the volatilized hydrocarbons with condensation means, and noncondensible hydrocarbons such as methane and other hydrocarbon gases which are not recoverable by ordinary condensation means. Condensible hydrocarbons can be separated and recovered by conventional means such as venturi scrubbers, indirect heat exchangers, wash towers, and the like. The undesirable gaseous products can be removed from the noncondensible hydrocarbons by means such as chemical scrubbing. Remaining uncondensed hydrocarbons can be sold as a product gas stream and can be utilized as the carrier gas for carrying the carbonaceous material to the pyrolysis reaction zone.

The particulate source of heat is formed in the combustion unit 50. The solids separated in the first cyclone separator 22 are passed down through the dipleg 78 into the fluidized bed 60 containing spent particulate source of heat and the carbon-containing solid residue of pyrolysis. As the solids drop down through the dipleg, hydrocarbons on the surface of the solids are stripped by an upward flow of stripping gas, nondeleteriously reactive with respect to pyrolysis products, such as steam. The stripping gas is introduced through gas inlets 120 on the side of the dipleg. The bed 60 is maintained in a fluidized state by an upward flow of fluidizing gas stream 91 into the vessel 52 through the gas inlet 62 and distributed by the distributor plate 64. The fluidizing gas can be nonreactive with respect to the solids in the fluidized bed, being for instance the off gas product of pyrolysis, or the gas may contain a portion of the oxygen required for oxidizing the solids to form the particulate source of heat.

A transport gas is introduced upwardly through the gas inlet 56 and nozzle 58 into the conduit 54. The transport gas preferably contains free oxygen. Other reactants which lead to the formation of carbon monoxide may be present. These include steam and carbon dioxide. When steam is present, hydrogen also is formed.

In the preferred process, the transport gas contains, as indicated, some oxygen to generate a portion of the heat necessary to raise the solids to the temperature required for feed to the pyrolysis reactor in the first combustion zone. However, the amount of oxygen is limited for if there is too much oxygen in the transport gas, the carbon monoxide generated in the transport line cannot be converted to carbon dioxide in the second combustion zone without introducing so much additional oxygen to the second combustion zone that the solids would be raised to a temperature above the temperature required for feed to the pyrolysis reactor.

As indicated in Figure 1, the transport gas can be an air stream 90 introduced upwardly through the gas inlet 56 and nozzle 58 into the conduit 54. A sufficient supply of this air stream at an appropriate oxygen content is maintained:

(1) to educt solids from the fluidized bed into the conduit; (2) to oxidize a portion of the carbon in the solids to heat the solids in the conduit and riser; and (3) to transport the solids and combustion products, including carbon monoxide, of the solids upwardly through the vertical riser 68 into the second combustion zone chamber 70. The fluidizing gas stream 91 passes through the annular gap or space 74 between the upper edge 72 of the conduit and the vertical riser 68 to help carry the solids upwardly into the second combustion chamber 70. If the top portion 71 of the conduit 54 is smaller in diameter than the riser 68, the flow of gas and solids upwardly into the riser from the conduit can serve to educt the fluidizing gas into the riser through the annular gap 74.

The velocity of the transport gas is maintained sufficiently high to educt solids into the conduit and convey them into the second combustion zone. For example, when the transport gas contains air as a source of oxygen, a diluent gas substantially free of free oxygen, e.g. nitrogen or flue gas, can be combined with the air to provide an oxygen-lean transport gas having sufficient velocity to educt and transport the solids without introducing too much oxygen to generate too much carbon monoxide. By diluting the heated air stream transport gas stream containing less than 20% oxygen by volume is formed.

The amount of oxygen in the transport gas is controlled to maintain the desired temperature in the riser. This is always less than the stoichiometric amount required to completely oxidize the carbon content of the solids. Because of this deficiency of oxygen and the relatively high temperature in the riser, which can range up to about 1100°F. (595°C.) in the case of the pyrolysis reaction zone maintained at a temperature in the region of 2000°F. (1100°C.) for a pyrolysis reaction designed to enhance gasification, appreciable amounts of carbon monoxide are formed.

Also, as the solids and combustion gases pass upwardly through the riser 68, carbon dioxide introduced in the transport gas and carbon dioxide formed by oxidation of carbon tends to react with additional carbon in the solids to form carbon monoxide according to the reaction:

$$C + CO_2 \rightarrow 2CO$$

Thus generally, less than half, and usually from 20 to 50% of the oxygen required to form the particulate source of heat is in the transport gas. The remainder of the oxygen required is introduced into the second combustion zone to oxidize the carbon monoxide from the first combustion zone to carbon dioxide.

Excess solids in the fluidized bed beyond what is required for oxidation to form the particulate source of heat represent the net solid product of the pyrolysis reaction, and are withdrawn from the vessel 52 through line 94.

The configuration of the combustion unit shown in the drawings and described above has many advantages. Among these is instant ignition of the solids entering the fluidized bed 60. When exposed to a source of oxygen the carbon in the carbon-containing solid residue is readily oxidized. If the carbon-containing solid residue has poor ignition properties oxygen can be introduced with the fluidizing gas to oxidize carbon in the solids in the fluidized bed to raise the temperature of the fluidized bed. During startup, a fuel gas followed by air can be utilized as a fluidizing gas to elevate the temperature of the solids in the fluidized bed above the solids ignition temperature.

Another advantage of the scheme shown in the drawings and described above is that the temperature in the first combustion chamber is easily controlled by controlling the amount of oxygen fed to the fluidized bed in the fluidizing gas stream 90.

Another advantage results from the large mass of solids in the fluidized bed. Because of this large mass, minor system upsets are damped by changes in the level of the fluidized bed. If the level in the fluidized bed rises, additional solids are removed through the withdrawal line 94 and additional solids are educted by the transport gas because of the higher differential pressure of the solids due to the increase in height of the bed. Conversely, if the level in the bed falls, fewer solids are withdrawn as product and less solids are educted by the transport gas because the differential pressure of the bed decreases. If any additional controls on the level of the fluidized bed are required, the jet flow of the source of oxygen can be varied. Thus the fluidized bed is a self-compensating system.

Another advantage of the configuration of the first combustion chamber and vessel is that because the solids are fluidized in the fluidized bed, withdrawal of solid product is facilitated. As the level of the solids in the fluidized bed rises, more solids are automatically withdrawn through the solids outlet line 94. This line extends upwardly into the vessel 52 and its height determines the average top 80 of the fluidized bed in the vessel.

A major advantage of the scheme shown in the drawings is that it provides a comparatively "fail-safe" method of preventing oxygen in the combustion unit 50 from entering the pyrolysis section 8. The height of the fluidized bed acts as a barrier against the backflow of oxygen through the dipleg 78 into the pyrolysis reactor. In addition, automatic control means can be provided to sense the level of the fluidized bed, and if the level drops too low, automatically to cut off the flow of the source of oxygen into the first combustion zone.

A source of oxygen is introduced through the gas inlet 82 into the second combustion zone. The amount of free oxygen introduced into the second combustion zone equals at least 50% of the molar amount of carbon monoxide entering the stage to completely oxidize carbon monoxide generated in the first combustion zone so the total potential heating value of the carbon oxidized in the first combustion zone is obtained. In addition, oxygen above the stoichiometric amount can be added to react with the carbon in the solids to heat the solids to the temperature required to form the particulate source of heat for introduction into the pyrolysis zone. The total oxygen feed to the two oxidation stages is at all times sufficient to raise the solids to the temperature required for feed to the pyrolysis zone. Typically the particulate source of heat has a temperature from about 100 to 500°F. (55 - 280°C.) higher than the pyrolysis zone temperature.

Introducing oxygen to oxidize carbon in the solid residue in two combustion zones serves to obtain maximum heating value from solid residue by oxidation. When the solid residue is oxidized where there is less than stoichiometric amounts of oxygen and/or the residence time is long, then some of the carbon dioxide in the reaction product gases tends to react with carbon in the solid residue to produce carbon monoxide. This is undesirable because more of the valuable carbon-containing solids residue has to be burned to achieve desired temperatures than if carbon dioxide were the only product. Net carbon monoxide formed is minimized and the carbon dioxide to carbon monoxide ratio maximized to maximize the amount of heat generated per unit free carbon combusted by using two combustion zones.

The formed particulate source of heat and the gaseous combustion products of the solids, as well as nonreactive components of the source of oxygen such as nitrogen, pass from the second combustion chamber to a second cyclone separator 84. In the separator the particulate source of heat is separated from the combustion gases for feed to the pyrolysis reaction zone. The gases 100 are discharged through the top of the cyclone 84. Because most of the carbon monoxide formed in the riser and conduit is oxidized to carbon dioxide in the second combustion zone, the combustion gases can be directly released to the atmosphere. However, if there are appreciable amounts of carbon monoxide or other pollutants in the combustion gas stream 100 from the second cyclone separator 84, these gases can be treated as by chemical scrubbing before release to the atmosphere.

Although the drawings show the second combustion zone and the second cyclone separation zone constituted by separate apparatus, it is possible to form the particulate source of heat from the preheated solids and separate the particulate source of heat from the gaseous combustion products simultaneously in a single cyclone oxidation-separation zone.

This modification has significant advantages. Among these advantages are reduced capital and operating costs for the process because a separator and a combustion zone are replaced with a single cyclone separator. In addition, production of carbon monoxide is minimized because short reaction times, which favour production of carbon dioxide, are obtained by using a cyclone vessel for oxidizing the carbon-containing solid residue. It is preferred that the residence times of solids in a cyclone oxidation-separation zone be less than 5 seconds, and more preferably, from 0.1 to 3 seconds. This short residence time favours production of carbon dioxide compared to carbon monoxide.

Another advantage of using a cyclone oxidation-separation zone is that carbon-containing solid residue fines, which are less valuable than larger particles are burned preferentially because of the more efficient separation of the larger particles from the fines in the cyclone.

The formed particulate source of heat separated from the gases in the second cyclone separation zone is passed through the standpipe 86 to the fluidized chamber 32 surrounding the inlet to the pyrolysis reactor. The standpipe is fluidized by an aeration gas nondeleteriously reactive with respect to pyrolysis products. The aeration gas is introduced through the inlets 88 along the length of the standpipe.

Although the invention has been described in terms of certain preferred embodiments other embodiments will be apparent to those skilled in the art. For example, steam can be injected along with the carbon-containing solid residue to the fluidizing chamber 32 to react with the hot particulate source of heat to form hydrogen gas by water-gas shift reactions. The hydrogen so produced can hydrogenate the volatilized hydrocarbons resulting from the pyrolysis of the carbonaceous material to upgrade their value. In addition, one or more cyclones in series or parallel as required can be used to replace the cyclone separators 22, 84.

The advantage of using more than one cyclone in series is that a fines fraction of the carbon-containing solid residue and a fines fraction of the particulate source of heat can be removed from the bulk of the particles so that the amount of solids carried over with the vapor mixture to a product recovery operation is minimized.

WHAT WE CLAIM IS:

1. A process for pyrolysis of solid carbonaceous materials by heat transferred thereto by a particulate source of heat to yield a particulate carbon-containing solid residue as a product of pyrolysis, the particulate source of heat being formed by oxidizing at least a portion of the particulate carbon-containing solid residue, characterised in that the particulate source of heat is generated by transporting at least a portion of the particulate carbon-containing solid residue formed by pyrolysis of the solid carbonaceous material to a fluidized bed around a substantially vertically oriented, open conduit in open communication with a substantially vertically oriented riser, the conduit and riser comprising a first combustion zone; educting solid residue from the fluidized bed into the first combustion zone and transporting it therethrough to a second combustion zone by injecting a transport gas upwardly into the conduit, while introducing a gaseous source of oxygen into said combustion zones to oxidize carbon in the solid residue.

2. A process according to claim 1, in which the amount of oxygen introduced into the second combustion zone is equal to at least 50% of the molar amount of carbon monoxide produced in the first combustion zone.

3. A process according to claim 1 or 2, in which the said conduit is spaced apart from the said riser and the particulate carbon-containing solid residue is fluidized in the fluidized bed by an upward flow of a fluidizing gas that passes into the riser through the space between the riser and the conduit.

4. A process according to claim 1, 2 or 3, in which the fluidized bed is fluidized by a fluidizing gas containing oxygen.

5. A process according to any one of claims 1 to 4, in which the formed particulate source of heat and gaseous combustion products from said second combustion zone are separated from one another in a cyclone separation zone.

6. A process according to any one of claims 1 to 4, in which the said second combustion zone comprises a cyclone oxidation separation zone.

7. A process according to claim 6, in which the residence time in the cyclone oxidation-separation zone is less than 5 seconds.

8. A process according to claim 7, in which the residence time in the cyclone oxidation-separation zone is less than 3 seconds.

9. A process according to any one of claims 1 to 8, in which the solid carbonaceous material is subjected to flash pyrolysis by continuously transporting particulate solid carbonaceous material feed contained in a carrier gas which is substantially nondeleteriously reactive with respect to products of pyrolysis to a vertically oriented, descending flow pyrolysis reactor containing a pyrolysis zone operated at a temperature below about 2000°F. (1100°C.); feeding the particulate source of heat at a temperature above the pyrolysis temperature to the pyrolysis reactor at a rate sufficient to maintain said pyrolysis zone at the pyrolysis temperature; forming a turbulent mixture of the particulate source of heat, particulate solid carbonaceous material feed and the carrier gas to pyrolyze the solid carbonaceous material feed and yield a pyrolysis product stream containing as solids, the particulate source of heat and a particulate carbon-containing solid residue of pyrolysis, and a vapour mixture of carrier gas and pyrolytic vapours comprising hydrocarbons; and passing the pyrolysis product stream from the pyrolysis reactor to a first separation zone to separate solids from the vapour mixture.

10. A process according to claim 9, in which the first separation zone is a cyclone separation zone.

11. A process according to claim 9 or 10, in which the turbulent mixture in the pyrolysis reactor has a solids content ranging from 0.1 to 10% by volume based on the total volume of the stream, and a weight ratio of the particulate source of heat to the solid carbonaceous material ranging from 2:1 to 20:1.

12. A process according to claim 9, 10 or 11, in which the pyrolysis zone is operated at a temperature above 600°F. (315°C.).

13. A process according to claim 12, in which the pyrolysis zone is operated at a temperature in the range 900 to 1400°F. (480 - 760°C.).

14. A process according to any one of claims 9 to 13, wherein the pyrolysis reactor has a solids feed inlet for the solid carbonaceous material feed and a vertically oriented chamber surrounding the upper portion of the pyrolysis reactor, the inner peripheral wall of said chamber forming an overflow weir to a vertically oriented mixing zone of the reactor, the particulate solid carbonaceous material being transported in a carrier gas to the solids feed inlet and thence being injected into the mixing zone, the particulate source of heat being fed

to the said vertically oriented chamber and being fluidized in such chamber by a flow of a fluidizing gas substantially non-deleteriously reactive with respect to the products of pyrolysis, the fluidized particulate source of heat discharging over said weir and downwardly into said mixing zone to form a turbulent mixture with the particulate solid carbonaceous material, said mixture passing downwardly from the mixing zone to the pyrolysis zone of the pyrolysis reactor to pyrolyze the solid carbonaceous material.

15. A process according to any one of claims 9 to 14, in which the residence time of the carrier gas in the pyrolysis zone of the pyrolysis reactor and in the first separation zone is less than 5 seconds.

16. A process according to claim 15, in which the said residence time of the carrier gas in the pyrolysis zone of the pyrolysis reactor and in the first separation zone is less than 3 seconds.

17. A process according to claim 16, in which the said residence time is in the range of 0.1 to 3 seconds.

18. A process according to any one of claims 9 to 17, in which the carbonaceous material is particles having a maximum dimension less than 1000 microns.

19. A process according to claim 18, in which the solid carbonaceous material is an agglomerative coal in the form of particles having a maximum dimension less than 250 microns.

20. An apparatus for forming a particulate solid source of heat from a particulate carbon-containing solid residue of pyrolysis of a carbonaceous material for pyrolysis of the solid carbonaceous material, comprising a vessel for containing a fluidized bed of said particulate carbon-containing solid residue of pyrolysis around an open, substantially vertically oriented conduit; a substantially vertically oriented riser in open communication with the conduit, said conduit and said riser comprising a first combustion chamber; a second combustion chamber in communication with the riser; means for introducing particulate carbon-containing solid residue of pyrolysis into the fluidized bed; means for injecting a transport gas upwardly into the conduit to educt carbon-containing residue from the fluidized bed upwardly into the combustion chambers to oxidise carbon in the said solid residue to form the particulate source of heat.

21. An apparatus according to claim 20, in which the conduit is separated from the vertical riser.

22. An apparatus for pyrolysis of solid carbonaceous materials, comprising a descending flow pyrolysis reactor; means for forming a turbulent mixture of a particulate source of heat and a solid carbonaceous material contained in a carrier gas for introduction into the pyrolysis reactor to pyrolyze the solid carbonaceous feed to form a pyrolysis product stream containing a vapour mixture and, as solids, the particulate source of heat and a particulate carbon-containing solid residue of pyrolysis; means for transferring the pyrolysis product stream from the pyrolysis reactor to a first separator adapted for separating solids from the vapour mixture in the pyrolysis product stream; means for passing the separated solids from the first separator to a vessel for containing a fluidized bed of the separated solids around an open, substantially vertically oriented conduit, a substantially vertically oriented riser being in open communication with the conduit and comprising therewith a first combustion chamber; a second combustion chamber in communication with the riser; means for injecting a transport gas upwardly into the conduit to educt solids from the fluidized bed upwardly into the conduit, to transport the solids through the first combustion chamber to the second combustion chamber; and means for introducing oxygen into the combustion chambers to oxidize carbon in said solids thereby to heat the solids to form the particulate source of heat and to produce gaseous combustion products; means for transferring the particulate source of heat from the second combustion chamber to a second separator adapted for separating the particulate source of heat from the gaseous combustion products; and means for transferring the separated particulate source of heat from the second separator to the pyrolysis reactor.

23. Apparatus according to claim 22, in which the conduit is spaced apart from the riser.

24. Apparatus according to claim 22 or 23, in which the first separator is a cyclone separator.

25. Apparatus according to claim 22, 23 or 24, in which the second separator is a cyclone separator.

26. Apparatus according to any one of claims 22 to 25, in which the pyrolysis reactor contains a substantially vertically oriented mixing section and a substantially vertically oriented pyrolysis section, and has a solids feed inlet, a substantially vertically oriented chamber surrounding the upper portion of the reactor and having an inner peripheral wall that forms an overflow weir to the vertically oriented mixing section, and the means for forming a turbulent mixture comprises means for feeding particulate source of heat to the vertically oriented chamber; means for introducing a fluidizing gas into the chamber to maintain the particulate source of heat therein in a fluidized state; and means for injecting the

solid carbonaceous feed contained in the carrier gas from the solids feed inlet into the mixing section to form the resultant turbulent mixture.

27. An apparatus for pyrolysis of agglomerative coals, comprising a descending flow pyrolysis reactor containing a substantially vertically oriented mixing section, a substantially vertically oriented pyrolysis section, a solids feed inlet, and a substantially vertically oriented chamber surrounding the upper portion of the reactor, the inner peripheral wall of the chamber forming an overflow weir to the mixing section, wherein an agglomerative coal feed contained in a carrier gas is combined with a particulate source of heat under turbulent flow conditions in the pyrolysis section of the pyrolysis reactor to yield a pyrolysis product stream containing, as solids, the particulate source of heat and a particulate carbon-containing solid residue of pyrolysis, and a vapour mixture; means for feeding the particulate source of heat to the vertically oriented chamber; means for introducing a fluidizing gas into the chamber to maintain the particulate source of heat therein in a fluidized state; means for passing a coal feed from the solids feed inlet into the mixing section; a first cyclone separator in communication with the pyrolysis reactor for separating solids in the pyrolysis product stream from the vapour mixture in that stream; a vessel for containing a fluidized bed of the separated solids around an open, substantially vertically oriented conduit, a substantially vertically oriented riser being in open communication with the vertically oriented conduit and separated therefrom, said conduit and riser comprising a first combustion chamber; a second combustion chamber in communication with the riser; means for injecting a transport gas upwardly into the conduit to educt solids from the fluidized bed upwardly into the conduit and to transport said solids through the first combustion chamber to the second combustion chamber; means for introducing oxygen into said combustion chambers to oxidize carbon in the solids to heat the solids and to form the particulate source of heat with attendant formation of combustion products; a dipleg from the first cyclone separator to the fluidized bed for transferring the separated solids from the first cyclone separator in communication with the second combustion chamber for separating the particulate source of heat from the gaseous combustion products; and a dipleg from the second cyclone separator to the chamber surrounding the upper portion of the pyrolysis reactor for transferring the particulate source of heat to the pyrolysis reactor.

28. Apparatus according to claim 20, 22 or 27, substantially as described with reference to and as shown in the accompanying drawings.

29. A process according to claim 1, substantially as described with reference to the accompanying drawings.

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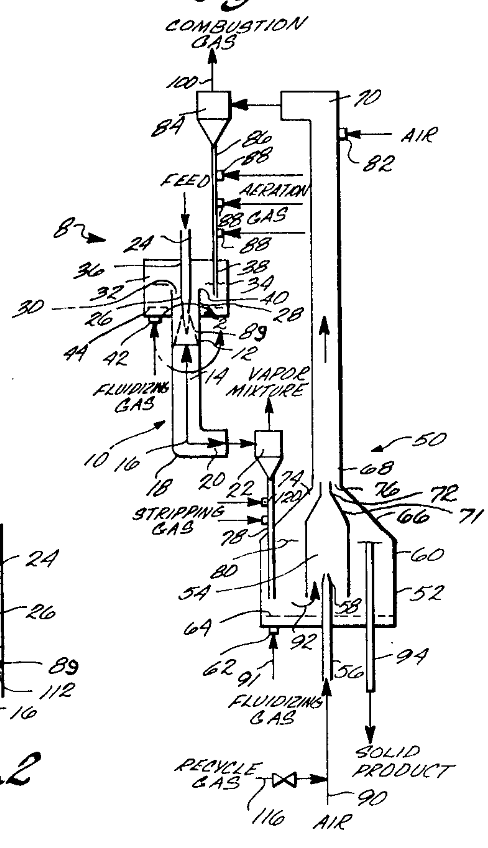
1583890 COMPLETE SPECIFICATION

1 SHEET *This drawing is a reproduction of the Original on a reduced scale*

Fig. 1



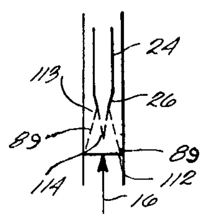


Fig. 2