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(54) FLUID-WALL REACTORS AND THEIR UTILIZATION IN HIGH TEMPERATURE CHEMICAL REACTION PROCESSES

(71) We, THAGARD TECHNOLOGY COMPANY, a limited partnership organised and existing under the laws of the State of California, of 2712 Kelvin Avenue, Irvine, State of California, 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 present invention relates to fluid-wall reactors for high temperature chemical reaction processes, as well as to the various processes which may be conducted in such reactors, many of which processes previously have been impractical or only theoretically possible. Both the fluid-wall reactor and the processes employed in such reactors utilize radiation coupling as a heat source, maintain the contemplated chemical reactions in isolation within a protective fluid blanket or envelope out of contact with the containing surfaces of the reactor, and provide a heat shield which substantially encloses the radiant energy heating means and the reaction zone to minimise the escape of radiation. The material from which the heat shield is fabricated functions as an insulator, inhibiting the transfer of heat from its interior to its exterior, and must be able to withstand the temperatures generated by the radiation coupling heat source.

The subject matter of the present invention is closely related to that of our co-pending application No. 42860/77 (Serial No. 1,530,315).

High temperature reactors are presently employed to carry out pyrolysis, thermolysis, dissociation, decomposition and combustion reactions of both organic

and inorganic compounds. Substantially all such reactors transfer heat to the reactants by conversion and/or conduction, but this characteristic inherently produces two major problems which limit the nature and scope of the reactions which may be carried out. Both problems result from the fact that in a conventional reactor which transfers heat to the reactants by convection, the highest temperature in the system is necessarily at the interface between the inside wall of the reactor and the reactant stream.

The first problem involves the limitations on available temperatures of reaction which are imposed by the strength at elevated temperatures of known reactor wall materials. The decreasing capability of such materials to maintain their integrity under conditions of increasing temperature is, of course, well known. However, since it is necessary that such materials be heated in order that thermal energy may be transferred to the reactant stream, available reaction temperatures have been limited by the temperature to which the reactor wall could be safely heated. This factor is particularly critical in cases where the contemplated reaction either must take place at or produces high pressures.

The second problem inherently results both because the wall of a conventional reactor is at the highest temperature in the system and because convective/conductive heat transfer requires contact between the wall and the reactant stream. Being at such elevated temperature, the reactor wall is an ideal if not the most desirable reaction site in the system and, in many instances, reaction products will accumulate and build up on the wall. Such build-up impairs the ability of the system to transfer heat to

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the reactants and this ever increasing thermal impedance requires the source temperature to be raised progressively just to maintain the initial rate of heat transfer into the reactant stream. Obviously, as the build-up increases, the required source temperature will eventually exceed the capabilities of the reactor wall material. Moreover, as additional energy is required to sustain the reaction, the process becomes less efficient in both the technical and economic sense. Thus, at the point where the contemplated reaction can no longer be sustained on the basis of either heat transfer, strength of materials, or economic considerations, the system must be shut down and cleaned.

Usually, cleaning is performed mechanically by scraping the reactor wall or chemically by burning off the deposits. In some continuous processes, it has been attempted to scrape the reactor wall while the reaction proceeds. However, the scraping tool itself necessarily gets hot, becomes a reaction site and, thereafter, must be cleaned. In any event, this down time represents a substantial economic loss. In many instances, a second system will be installed in order to minimize lost production time. However, such additional equipment generally represents a substantial capital investment. Some high temperature chemical reactors include a tube which is heated to a temperature at which its inner walls emit sufficient radiant energy to initiate and sustain the reaction. However, as in the case of conductive and convective reactors, for reactions yielding solid products there is frequently an undesirable build-up of product on the tube walls which leads to reduced heat transfer and even clogging of the tube.

The reactor disclosed in U.S. Patent No. 2,926,073 is designed to produce carbon black and hydrogen by the pyrolysis of natural gas. The process is stated to be continuous but, in practice, the convective heat transfer principle under which the reactor operates causes serious problems both in sustaining and controlling the reaction. Since the heated tubes of the reactor are ideal reaction sites, carbon invariably builds up and eventually clogs the system. More serious, however, is the problem of thermal runaway which can result in explosions. With respect to this condition, it has been determined that during pyrolysis of natural gas, thermal conductivity of the gas phase suddenly increases from about five to thirty times, depending upon the composition of the gas. Because the temperatures in a conventional convective reactor cannot be regulated with sufficient speed and accuracy to compensate for this phenomenon, in some

instances the system would become unstable and explosions would result. Such conditions are inherent in conventional reactors and, as yet, no way has been found to overcome this problem.

U.S. Patent No. 3,565,766 represents a recent attempt to upgrade coal by pyrolysis. The disclosed system comprises a series of hollow steel vessels which act as a multi-stage fluidized beds at successively increasing temperatures up to about 1600°F. Fluidization at lower temperatures is achieved by an inert gas which may itself supply heat although external heating is contemplated. At higher temperatures, fluidization is achieved by the overhead gas obtained in the final stage; and, in the final stage, temperature is maintained by internal combustion of the char in air or oxygen. Because it relies primarily upon heat transfer by convection, this system is subject to many of the defects and disadvantages which have previously been discussed.

The apparatus for the manufacture of carbon black disclosed in U.S. Patent No. 2,062,358 includes a porous tube disposed within a heating chamber. Hot gas is directed from a remote furnace into the chamber, and thereafter forced through the wall of the porous tube to mix with the reactants. Thus, only convective transfer of heat from a fluid to reactants is employed. This necessitates the flow of a large volume of fluid through the heating chamber in order to make up for heat losses.

U.S. Patent No. 2,769,772 discloses a reactor for heat-treating fluid materials such as hydrocarbons which includes two concentric tubes disposed in a flame heated furnace. Reactants flow axially through the pervious inner concentric tube. A heat-carrier gas flowing in the annular chamber between the concentric tubes is heated by contact with the outer wall. Fluids in the inner tube are heated by convection when the heat-carrier gas passes through the pervious wall and mixes with them. Radiant heat transfer is expressly avoided. In fact, it is impossible to heat the inner tube without simultaneously heating the outer tube to at least as high a temperature.

The surface-combustion cracking furnace of U.S. Patent No. 2,436,282 employs the convective heat carrier gas principle similar to that of U.S. Patent No. 2,769,772. The furnace includes a porous, refractory tube enclosed by a jacket. A combustible fluid from an annular chamber is forced through the porous wall to the inside of the tube where it is ignited. It is evident, however, that the combustible fluid in the annular chamber will explode unless it is forced through the porous wall at a rate faster than the rate of flame

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propagation back through the wall. Likewise, the temperature in the annular chamber must be maintained below the ignition temperature of the gas/air mixture. Combustion products from the surface flame mix with reactants in the furnace diluting and possibly reacting with them. Heat is imparted to the reactances by convective mixing of the combustion products and the reactants.

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U.S. Patents Nos. 2,670,272; 2,670,275; 2,750,260; 2,915,367; 2,957,753; and 3,499,730 disclose combustion chambers for producing pigmentary titanium dioxide by burning titanium tetrachloride in oxygen. In the '275 patent which is representative of this group, titanium tetrachloride is burned in a porous, refractory tube. An inert gas is continuously diffused through the porous tube into a combustion chamber where it forms a protective blanket on the inner surface of the tube. This gaseous blanket substantially reduces the tendency of the titanium dioxide particles to adhere to the walls of the reactor. Since the combustion of titanium tetrachloride is an exothermic reaction, no provision is made to supply heat to the reaction mixture as it passes through the tube. In fact, the '275 patent teaches that it is advantageous to remove heat from the reactor chamber either by exposing the porous tube assembly to the atmosphere or by circulating a cooling fluid through a coil disposed about the porous tube.

The present invention provides a process for carrying out a chemical reaction at an elevated temperature, wherein radiation is caused to be incident on one or more reactants situated in a reaction zone defined by a wall of fluid substantially transparent to that radiation, the wall of fluid being located within a heat shield of refractory material which reflects radiation, sufficient radiant energy being absorbed in the reaction zone to raise the temperature of or at least one reactant to sustain the chemical reaction.

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The fluid wall defining the reaction zone is preferably in the form of an envelope of generally annular cross-section situated within a reactor tube having an inlet end and an outlet end, the reactor tube being positioned within the heat shield.

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The invention further provides a reactor for carrying out a chemical reaction at an elevated temperature, comprising a reactor tube having an inlet end and an outlet end, means for generating an envelope of fluid to define a fluid-walled reaction zone within the reactor tube, and means for causing in operation, sufficient radiant energy to be incident upon the reaction zone to raise the temperature of one or

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more reactants situated there to sustain a chemical reaction, the reactor tube being located within a heat shield of refractory material capable of reflecting the said radiant energy.

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The process of the invention is advantageously carried out as follows. An annular envelope of an inert fluid which is substantially transparent to radiation is generated, the envelope having a substantial axial length. Next, at least one reactant is passed through the core of the envelope along a predetermined path which is substantially coincident with the envelope axis, the reactants being confined within the envelope. After the reactant flow has started, high intensity radiant energy is directed through the envelope to coincide with at least a portion of the path of the reactants. Sufficient radiant energy is absorbed in the core to raise the temperature of the reactants to a level required to sustain the desired chemical reaction.

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As used herein, the terms "radiant energy" and "radiation" are intended to encompass all forms of radiation including high-energy or impacting nuclear particles. However, because the practical use of such radiation is not possible under the present state of the art electromagnetic radiation, particularly of wavelengths ranging from 100 microns to 0.1 microns, is considered to be the primary energy source upon which design considerations are to be based.

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In the event that the reactants are themselves transparent to radiant energy, an absorptive target may be introduced into the reactant stream, or introduced prior to the introduction of the reactants. The target will absorb sufficient radiant energy to raise the temperature in the core to the desired level. In some instances, however, while the reactants are transparent to radiation, one or more of the reaction products will be an absorber. In such event, once the reaction has been initiated the target may be withdrawn and the reaction will continue. An example of such reaction is the pyrolysis of methane to carbon and hydrogen.

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Some reactions will reverse either partially or completely if the reaction products are not cooled immediately and rapidly. In such cases, it is further contemplated that cooling of reaction products and any remaining targets to prevent such undesired chemical reactions be carried out immediately upon completion of the desired reaction.

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The high temperature fluid-wall reactor of the present invention transfers substantially all of the required heat to the reactants by radiation coupling either

directly or via a target. As previously mentioned, the reactor preferably includes a tube having an inlet and an outlet end, the interior of the tube defining a reaction zone. In this preferred embodiment, means for introducing an inert fluid into the reactor chamber provide a protective blanket for the radially inward surface of the reactor tube and means for introducing at least one reactant into the reactor tube through the inlet and cause such reactants to be directed in a predetermined path axially of the reactor tube. The inert fluid blanket confines the reactants substantially centrally within the reactor chamber and out of contact with the reactor tube walls. High intensity radiant energy is generated and directed into the reactor tube to coincide with at least a portion of the path of the reactants, sufficient radiant energy being absorbed to raise the temperature of the reactants to a level required to sustain the desired chemical reaction.

The reactor tube is advantageously made of a fabric of a fibrous refractory material capable of emitting sufficient radiant energy to raise the temperature of reactants within the reaction zone to a level required to sustain the desired chemical reaction. Such a fabric has a multiplicity of pores of such diameter as to permit a uniform flow of sufficient inert fluid which is substantially transparent to radiant energy through the tube wall to constitute a protective blanket for the radially inward surface of the reactor tube. In this especially preferred embodiment, a fluid-tight, tubular pressure vessel encloses the reactor tube to define an inert fluid plenum between the reactor tube and the pressure vessel, the inlet and outlet ends of the reactor tube being sealed from the plenum. The pressure vessel has at least one inlet for admitting the inert fluid which is directed under pressure into the plenum and through the porous tube wall into the reaction zone. This preferred reactor further includes means for introducing at least one reactant into the reaction zone through the inlet end of the reactor tube. Thereafter, the reactants are directed in a predetermined path axially of the reactor tube and are confined by the protective blanket substantially centrally within the reaction zone and out of contact with the inner wall of the reactor tube. At least one electrical heating element is disposed within the plenum and spaced radially outwardly of the reactor tube for heating the reactor tube to the temperature level at which it emits sufficient radiant energy to initiate and sustain the desired chemical reaction. The radiant energy is directed into the reaction zone substantially coincident with at least a portion of the

path of the reactants. The heat shield mentioned above is disposed within the pressure vessel substantially enclosing the heating elements and the reaction zone. The heat shield reflects radiant energy inwardly toward the reaction zone.

In contrast to the conventional convective reactors, the present invention relies upon radiation coupling to transfer heat to the reactant stream. The amount of heat transferred is independent both of physical contact between the reactor wall and the stream and of the degree of turbulent mixing in the stream. The primary consideration for heat transfer in the present system is the radiation absorption coefficient (α) of the reactants. The inert fluid blanket which protects the reactor wall is substantially transparent to radiation and thus exhibits a very low value of (α). This enables radiant energy to be transferred through the blanket to the reactant stream with little or no energy losses. Ideally, either the reactants themselves or a target medium will exhibit high (α) values and will thus absorb large amounts of energy, or alternatively, the reactants may be finely divided (as in a fog) such that the radiation is absorbed by being trapped between the particles. Since materials which are good absorbers are generally good emitters of radiation, when the reactants or targets are raised to a sufficiently high temperature, they become secondary radiators which re-radiate energy throughout the entire reacting volume and further enhance the heat transfer characteristics of the system. This occurs almost instantaneously and is subject to precise and rapid control. Moreover, the re-radiation phenomenon which ensures rapid and uniform heating of the reactants is completely independent of the degree of turbulent mixing which may exist in the reactant stream.

The present high temperature chemical process and apparatus provide a solution to problems which have plagued the art and thus permit the carrying out of reactions which heretofore have been impractical or only theoretically possible. Because heat is supplied by radiation coupling rather than by convection and/or conduction, the temperature of the reactant stream may be independent of both the temperature of the reactor wall and of the condition of the reactant stream, and the serious strength of materials problem is overcome. Two embodiments of the present reactor, described below in more detail, provide a heated wall as a source of radiant energy, but the heated wall is not subjected to the high pressures which are normally attendant to many kinds of reactions. For this reason, refractory materials such as

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carbon or thorium oxide, which are not
suitable for use as a wall material in a
conventional reactor, may be successfully
employed. As compared to the most
5 temperature-resistant alloys which melt at
about 2900°F., thorium oxide, for example,
is serviceable at temperatures greater than
5400°F. This feature permits reaction
temperatures far in excess of those
10 presently achievable and reactions which
had been only theoretically feasible may be
carried out. In the preferred embodiment
of the invention wherein the reaction is
carried out in a reactor tube, the
15 temperature of the reactor tube is
preferably at least 2300°F.
Carbon cloth, the preferred porous
refractory material for the reactor tube, is
relatively inexpensive, readily available,
and may be formed into reactor tubes
20 substantially larger than those of cast
porous carbon presently available. Since
carbon cloth is normally flexible, any
attempt to force an inert gas radially
inwardly through a reactor tube of such
25 material would ordinarily cause the tube to
collapse. Accordingly, the present
invention contemplates the depositing of a
layer of pyrolytic graphite on the cloth to
stiffen it sufficiently to withstand the
30 pressure differential maintained between
the inert fluid plenum and the reaction
zone. Depositing a layer of pyrolytic
graphite on the cloth also permits control of
the porosity of the fabric.
35 The provision of the protective inert fluid
blanket, which is made possible largely by
the use of radiation coupling, isolates the
reactor wall from the reactant stream and
makes it impossible under normal operating
40 conditions for any precipitates or other
deposits to accumulate and clog the system.
In the event that a corrosive blanket fluid
such as steam is to be used, surfaces of the
reactor tube, heating elements and heat
45 shield which are maintained at high
temperatures and in contact with the
blanket gas when the reactor is in operation
may be coated with a thin layer of
refractory oxide such as thorium oxide,
50 magnesium oxide, or zirconium oxide. The
refractory oxide may be deposited on these
surfaces by heating the reactor to above the
dissociation temperature of a volatile metal-
containing compound, introducing this
55 compound into the reactor chamber and
allowing it to dissociate, depositing a layer
of metal on the heated surfaces. Thereafter,
a gas or other suitable material such as
molecular oxygen may be introduced into
60 the reactor chamber to oxidize the metal
layer, forming the desired refractory oxide.
Alternatively, the refractory coating may
be achieved in a single step if a volatile
65 metal-containing compound which

pyrolyzes directly to an oxide is employed
as a refractory deposition agent.
The use of radiation coupling further
enables the accurate and almost
70 instantaneous control of heat transfer rates
which is impossible to achieve in a
conventional convective reactor.
Furthermore, the present reactor may
provide a power flux at the reaction site in
excess of 10,000 watts/cm²; preferably the
75 power flux in the reaction zone is at least
180 watts/cm². Even this lower figure
represents a great improvement over the
2—3 watts/cm² which is ordinarily obtained
in conventional reactors. And, the use of a
80 heat shield to provide the containing
surface or surfaces of a zone within which
all reactions take place enables the
achievement of unusually favorable thermal
efficiencies.
85 The reactions which may be carried out
by the process of this invention as
implemented by the present reactor are
many and varied. For example, organic
compounds, particularly hydrocarbons,
90 may be pyrolyzed to produce carbon and
hydrogen without the attendant build-up
and thermal runaway problems which were
encountered in the prior art. Saturated
hydrocarbons may be partially pyrolyzed to
95 obtain unsaturated hydrocarbons; thus, for
example, propane and ethane may be
dehydrogenated to propylene and ethylene,
respectively. Unsaturated hydrocarbons
may be partially pyrolyzed in the presence
100 of hydrogen to form saturated
hydrocarbons and, more specifically,
petroleum products may be thermally
cracked. Thus, gas oil may be readily
converted into diesel oil, kerosene,
105 gasoline fractions or even methane.
Halogen intermediates may be added
to partially pyrolyzed hydrocarbons
to produce higher molecular weight
compounds. Hydrocarbons may be
110 completely or incompletely pyrolyzed in
the presence of steam to form carbon
monoxide and hydrogen; additional
hydrogen may then be added and the
reaction carried out to form alkane series
115 hydrocarbons which are high BTU-value
fuel gases.
Inorganic compounds may likewise be
pyrolyzed. For example, salts or oxides of
120 iron, mercury, silver, tungsten and
tantalum, among others, may be dissociated
to obtain pure metals. Oxides of iron,
nickel, cobalt, copper and silver, to name a
few, may be directly reduced in the
125 presence of hydrogen with the same
result. This list is by no means intended to
be exhaustive.
Novel composite products may also be
produced by the present process. For
130 example, carbon or talc particles coated

with silicon carbide may be obtained. This product serves as an excellent abrasive because as it is used, it continually breaks up and forms fresh new sharp surfaces. Particles of certain elements such as U^{235} may also be encapsulated in a chemically-tight envelope of another material such as carbon; this particular product is useful as a nuclear reactor fuel element.

It is further contemplated that the present invention may provide the terminal step in conventional aerobic incineration of waste such as garbage and sewage. The relatively low temperatures encountered in current incineration processing techniques permit the formation of organic peroxides and oxides of nitrogen which are major contributors to photochemical smog and other forms of air pollution. Because such compounds are not stable at the higher processing temperatures afforded by the present invention, a waste incineration effluent which is very low in pollutants may be obtained.

Further, the present invention contemplates the high temperature anaerobic destructive distillation and/or disassociation of waste to yield useful products such as carbon black, activated charcoal, hydrogen, and glass cullet, to name a few. The addition of steam to carbonaceous waste will produce carbon monoxide and hydrogen which may then be processed in the conventional manner to obtain fuel gases. Finally, the addition of hydrogen to carbonaceous waste will produce petroleum-equivalent heavy oils and other petroleum products. Thus, substantial reduction in air pollution as well as significant economic gains may be realized through such contemplated applications of the present invention.

The present invention represents a major breakthrough in the art. Because it makes available for the first time a source of thermal energy which has never been harnessed in this manner, its potential applications are numerous and varied. Moreover, in surmounting the strength of materials problem which has shackled the art for many years, this invention makes possible in the practical sense many useful chemical reactions which has long been known but which could not be performed because of temperature limitations inherent in reactors which depend upon convective and/or conductive heat transfer.

Figure 1A is an elevation in section of the inlet end of a first embodiment of the reactor of the present invention;

Figure 1B is an elevation in section of the outlet end of the first embodiment of the reactor of the present invention;

Figures 1A and 1B represent halves of an integral structure which has been

divided along line A—A in order to provide an illustration of sufficient size to show clearly certain structural details;

Figure 1C is a perspective in partial section of the first embodiment of the reactor of the present invention wherein certain elements have either been removed or illustrated diagrammatically to illustrate more clearly the operation of the reactor;

Figure 2 is a section taken substantially along line 2—2 of Figure 1A;

Figure 3 is a section taken substantially along line 3—3 of Figure 1B;

Figure 4 is a section taken substantially along line 4—4 of Figure 1A;

Figure 5 is a perspective of a portion of the reactor tube heating means of the first embodiment of the present invention;

Figures 6A, 6B, 6C and 6D together constitute a composite elevation in partial section of a second embodiment of the reactor of the present invention; the integral structure of the reactor has been divided along lines A—A, B—B and C—C, respectively, in order to provide an illustration of sufficient size to show clearly certain structural details;

Figure 7 is a section taken substantially along line 7—7 of Figure 6A;

Figure 8 is a section taken substantially along line 8—8 of Figure 6B;

Figure 9 is a section taken substantially along line 9—9 of Figure 6B;

Figure 10 is a section taken substantially along line 10—10 of Figure 6C;

Figure 11 is a section taken substantially along line 11—11 of Figure 6C;

Figure 12 is an elevation in section of a modified post-reaction treatment assembly for use in the second embodiment of the reactor of the present invention;

Figures 13A and 13B together constitute a composite elevation in partial section of a modified inlet assembly for use in the second embodiment of the present invention; the integral structure of the inlet assembly has been divided along line D—D in order to provide an illustration of sufficient size to show clearly certain structural details.

Figure 14 is an elevation/schematic view apparatus for pre-processing and introducing solid reactants into an inlet assembly as shown in Figures 13A and 13B.

Figure 15 is a schematic representation illustrating a refractory coating and etching system for use in the reactor of Figures 6A to 11 (the second embodiment);

Figure 16 is a schematic diagram of a temperature regulation circuit for use in the reactor of Figures 6A to 11 (the second embodiment).

Figure 17 is a graphical representation of the electrical resistance of a heating element of the reactor of Figures 6A to 11

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(the second embodiment) as a function of temperature and the number of layers of refractory fabric which constitute such element; and

5 Figure 18 is a schematic representation illustrating the operation of the several control systems of the reactor of Figures 6A to 11 (the second embodiment).

10 Referring to Figures 1A to 5 inclusive, and particularly to Figures 1A to 1C, a first embodiment of the present reactor 60 comprises a reactor tube 61 having an inlet end 62 and an outlet end 63; the interior of the tube 61 defines a reactor chamber 65.

15 The reactor tube 61 is made of a porous material which is capable of emitting radiant energy; preferably the pore diameter is in the range of about 0.001 to 0.020 inch to permit uniform flow of sufficient inert fluid through the tube wall to provide an adequate protective blanket.

20 Other wall constructions such as mesh, screening or various types of perforations may also be used to provide the desired result. The reactor tube 61 may be made from materials such as graphite or another form of carbon, sintered stainless steel, sintered tungsten, or sintered molybdenum, and, inorganic materials such as oxides of thorium, magnesium, zinc, aluminium or zirconium, among others. Tungsten, nickel and molybdenum are also suitable for use as mesh or screening.

25 A fluid-tight, tubular pressure vessel 70 which is preferably made of stainless steel encloses the reactor tube 61. The integrity of the vessel 70 is maintained by a series of sealing flanges 71, 72; 73, 74; and 75, 76 which join the several sections of the reactor 60. Flanges 72, 73 and 76 further are grooved to receive stainless steel O-rings 77, 78 and 79, respectively, which act as pressure seals. The reactor tube 61 is slidably mounted at one end within a graphite sleeve 81 which allows for any elongation of the tube 61 which may occur during operation at elevated temperatures.

30 The pressure vessel 70 further includes an inlet 83 for admitting an inert fluid, which is substantially transparent to radiation in that it has a low (α) value. Fluids which are suitable for this purpose include simple gases such as helium, neon, argon, krypton and xenon; complex gases which do not decompose to form a solid product such as hydrogen, nitrogen, oxygen and ammonia; and, liquid or gaseous water. The term "inert" as used herein, involves two factors: the ability of the fluid to react chemically with the material of the reactor tube 11 and the ability of the fluid to react chemically with the materials which are being processed. Thus, the selection of an "inert" blanket fluid depends in each

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instance upon the particular environment.

Except as otherwise specifically provided, it is desirable that the fluid be inert with respect to the reactor tube and it is usually desirable that the fluid be inert with respect to the reaction which is carried out. However, it is contemplated that in some instances the "inert" fluid of the protective blanket shall also participate in the reaction as, for example, where iron or carbon particles are reacted in the presence of a steam blanket to produce iron oxide and hydrogen or carbon monoxide and hydrogen, respectively.

The inert fluid is first directed under pressure into a plenum 85 which is defined between the reactor tube 61 and the pressure vessel wall 70. Thereafter, such fluid is directed through the porous wall of the tube 61 into the reactor chamber 65 to constitute a protective blanket for the radially inward surface of the reactor tube 61.

Means for cooling the pressure vessel 70 comprises cooling coils 87 which are disposed about the radially outward surface of the pressure vessel 70. The coils 87 are preferably covered with a flame-sprayed aluminium coating which enhances the thermal contact between the vessel 70 and the coils 87 to increase cooling efficiency. Such coils 87 are also disposed about a view-port 88 which is provided in the pressure vessel wall.

As shown best in Figures 1A and 2, the reactants are introduced into the reactor chamber 65 through the inlet end 62 of the reactor tube 61. Means for introducing the reactants comprises an inlet section 90 which is mounted in fluid-tight relationship by flanges 71, 72 adjacent the inlet end 62 of the tube 61. The reactants are carried in a gaseous stream through inlet 91, past a tangential baffle 92 and into a plenum 93 which is defined between an outer wall 94 and a diffuser 95. Suitable materials for the diffuser 95, whose function is to minimize turbulence in the stream, include porous carbon, steel wool and mesh screening. The reactants are directed in a predetermined path axially of the reactor tube 61 and are confined by the protective blanket substantially centrally within the reactor chamber 65 and out of contact with the inner wall of the reactor tube.

In this embodiment, the reactor tube 61 itself generates the high-intensity radiant energy which is directed centrally therewithin substantially coincident with at least a portion of the path of the reactants. Heating is provided by a plurality of carbon heating elements 100a—100f which are disposed radially outwardly of and spaced circumferentially about the tube 61; the heat generated by the elements 100 is transferred to the tube 61 by radiation. As

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best shown in Figures 2A, 5 and 6, electrodes 100a and 100b, for example, are embedded at one end in an arcuate carbon cross-over element 101a; electrodes 100c and 100d are embedded in cross-over 101b; and, electrodes 100e and 100f are likewise embedded in cross-over 101c. Tubular alumina spacers 102a and 102b (and a third one not shown) have the dual function of centering the porous reactor tube 61 and of dividing the three circuits. Referring specifically to Figures 1B and 3, each carbon heating element 100a—100f is mounted at its other end in a copper bus bar electrode 104. Although there are six such electrodes 104, only one is actually shown in Figure 3 as a matter of convenience. Each copper bus bar electrode 104 includes a phenolic flange 105 and a ceramic insulator 106. The electrode 104 is cooled by water which circulates in an internal channel 107, entering through inlet 108 and exiting through outlet 109. A high current electrical connection is illustrated at 110. A polytetrafluoroethylene seal 111 assists in preventing any leakage from the pressure vessel 70. The electrical system illustrated herein is particularly suitable for use with a three-phase power source. However, other systems may be used where circumstances warrant. It is further contemplated that the porous tube 61 may itself be heated directly by electrical resistance; in such event the electrodes 100 may be eliminated.

The thermal efficiency of the tube heating means is further improved by the provision of a molybdenum heat shield 120 which directs electromagnetic radiation from the carbon heating elements 100 toward the porous tube 61. In that the heat shield 120 reflects rather than transfers heat, it functions as an insulator and may thus be made of any material which exhibits this characteristic and which can withstand the temperatures generated by the heating elements 100. The heat shield 120 is disposed within the pressure vessel 70 radially outwardly of the heating elements 100 and preferably comprises a flat strip of rectangular cross-section which is wound in a series of helical turns. The inert blanket gas enters through the inlet 83 and can circulate freely throughout the plenum 85.

If the reactants themselves do not exhibit a relatively high radiation absorption coefficient (α), a radiant energy absorptive target must be introduced into the reactor chamber 65 coincident with at least one point along the path of the reactants on which radiation is incident. The target medium, which may be a finely divided solid such as carbon powder or other suitable material, is introduced into the reactor chamber 65 through an inlet 121

and absorbs sufficient radiant energy to raise the temperature of the reactants to the required level.

Alternatively, the target may be a liquid such as tar, asphalt, linseed oil or diesel oil, and may include solutions, dispersions, gels and suspensions of varied make-up which may be readily selected from available materials to suit particular requirements. The target may be a gas which preferably exhibits absorption in the electromagnetic spectrum from 100 microns to 0.01 microns; such gases include ethylene, propylene, oxides of nitrogen, bromide, chlorine, iodine, and ethyl bromide. The target may also be a solid element made of a material such as carbon which is disposed in the reactor chamber 65 along at least a part of the path of the reactants where the radiation is incident.

Other means for raising the temperature of the reaction to the required level may include an electrically heated element, an electric arc or a flame disposed within the reactor chamber 65 coincident with at least a part of the path of the reactants at or before the incidence of the radiation. In such instances, the initiating heat source is self-contained and is not derived from the radiant energy generating means. Such means are particularly useful where the reactants themselves are transparent to radiation but at least one of the reaction products is an absorber. Thus, once the contemplated reaction has been initiated, the temperature raising means may be deactivated because the reaction products will absorb sufficient radiant energy to sustain the reaction. Likewise if a target medium is used, it may be discontinued or withdrawn once the reaction has begun. An example of a reaction where a target or other initiating means is required only at the outset is the pyrolysis of methane to produce carbon and hydrogen.

As previously stated, some reactions will reverse either partially or completely if the reaction products are not cooled immediately and rapidly. For this purpose, reaction product cooling means 125 may be provided within the reactor chamber 65 adjacent the outlet end 63 of the reactor tube 61. The cooling means 125 is suitably disposed substantially centrally within the reactor chamber 65 and may, for example, include a tubular member having an internal channel through which is circulated a coolant such as water, the radially inward surface of the said tubular member being designed to constitute an absorber of radiant energy. As the reaction products, remaining reactants and targets, if any, pass within the cooled tube 125 heat is transferred rapidly by radiation coupling and the system is effectively quenched to

prevent any further undesired chemical reactions.

This embodiment has the major advantage that the inert fluid blanket is introduced into the chamber 65 in a radially inward direction. This is better than the possible alternative arrangement in which the blanket is introduced axially into the chamber. It will be appreciated that laminar flow can be maintained for only relatively short distances before turbulence causes intermixing and destroys the integrity of the protective blanket. Because radial blanket introduction does not require laminar flow of the blanket fluid, much greater axial reactor chamber lengths may be obtained. All that need be done in the described embodiment is to maintain the absolute level of the inert fluid pressure greater than the absolute level of the pressure in the reactant stream in order to prevent any reactants and/or reaction products from impinging upon the reactor tube 61. This feature aids in making this embodiment especially suitable for large scale commercial operation.

The reactor tube 61 must be heated and may operate at temperatures in excess of 5400°F. as in the case where porous thorium oxide is the base material. The hot wall 61 is not subject to a pressure gradient, except perhaps the relatively small differential between the fluid blanket and the reactive stream (the pressure difference required to maintain the desired rate of flow of the inert fluid through the wall of the reactor tube 61). The pressure is borne by the stainless steel pressure vessel wall 70 which, of course, is cooled by the coils 87 and thus is not subject to thermal stress. Accordingly, a refractory material, such as carbon or thorium oxide, which can withstand temperatures far in excess of those tolerable by conventional reactor wall materials but which are unsuitable for use in a conventional convective reactor, may now be employed for the first time to provide a practical, ultra-high-temperature system.

Referring to Figures 6A to 14 inclusive, a second embodiment of the present high temperature chemical reactor, which represents an improvement of the first embodiment, generally comprises an inlet assembly 200 and electrode assembly 300, a main assembly 400, and a post-reaction treatment assembly 500. The principal elements of this reactor include:

(1) A reactor tube 401 which has an inlet end 402 and an outlet end 403; at least a portion of the interior of the tube 401 defining a reaction zone 404. The reactor tube 401 is made of a fabric of a fibrous refractory material capable of emitting

temperature of reactants within the reaction zone 404 to a level required to initiate and sustain the desired chemical reaction. The fabric has a multiplicity of pores of such diameter as to permit a uniform flow of sufficient inert fluid which is substantially transparent to radiant energy through the tube wall to constitute a protective blanket for the radially inward surface of the reactor tube 401.

(2) A fluid-tight, tubular pressure vessel (which has an inlet assembly section 201, an electrode assembly section 301, a main assembly section 405, a post-reaction treatment assembly section 501) encloses the reactor tube 401 to define an inert fluid plenum 406 between the reactor tube 401 and the pressure vessel. The inlet and outlet ends, 402 and 403, respectively, of the reactor tube 401 are sealed from the plenum 406. The pressure vessel has a first inlet 408 and a second inlet 409 for admitting the inert fluid which is directed under pressure into the plenum 406 and through the porous tube wall 401 into the reaction zone 404.

(3) Means for introducing reactants, either gaseous, liquid, or solid, into the reaction zone 404 through the inlet end 402 of the reactor tube 401. The reactants are directed in a predetermined path axially of the reactor tube 401 and are confined by the protective blanket substantially centrally within the reaction zone 404 and out of contact with the inner wall of the reactor tube 401.

(4) Electrical means including heating elements 302a, 302b, and 302c (see Figure 10) which are disposed within the plenum 406 and spaced radially outwardly of the reactor tube 401 for heating the reactor tube to the temperature level at which it emits sufficient radiant energy to initiate and sustain the desired chemical reaction. The radiant energy is directed into the reaction zone 404 substantially coincident with at least a portion of the path of the reactants.

(5) A heat shield 410 which is disposed within the pressure vessel substantially enclosing the heating elements 302a, 302b, and 302c and the reaction zone 404. The heat shield 410 reflects radiant energy inwardly toward the reaction zone 404.

A. Inlet Assembly

Referring particularly to Figures 6A and 7, the pressure vessel inlet assembly section 201 is a tubular member having first and second flanges, 202 and 203, at its respective ends. An annular nozzle block 204 is secured to an annular sealing flange 205 which, in turn, is secured in fluid-tight relationship to the first flange 202 of the inlet assembly pressure vessel section 201.

A principal atomizing gas inlet tube 206 extends through the annular nozzle block 204 and is fixedly secured thereto by a support flange 207. An O-ring 209 in the support flange 207 assures a fluid-tight seal between the principal atomizing gas inlet tube 206 and the flange 207. An inlet fitting 210 is secured to an end of the principal atomizing gas inlet tube 206 as shown in Figure 6A. Atomizing gas enters a plenum 211 through inlet 212.

A principal liquid reactant inlet tube 214 is disposed within the principal atomizing gas inlet tube 206 and extends substantially coextensively therewith. A principal liquid reactant enters the tube 214 through inlet 215 in fitting 210.

As best shown in Figure 6B, a fogging nozzle 216 is secured to the outlet end of both the principal atomizing gas inlet tube 206 and the principal liquid reactant inlet tube 214. The fogging nozzle 216 includes a tubular shroud 217 which is secured to and disposed radially outwardly of the nozzle as shown. The axis of the shroud 217 is substantially parallel to the axis of the reactor tube 401. In operation, the liquid reactant and the atomizing gas are directed under pressure through tubes 214 and 206, respectively, and, under pressure, are mixed within the nozzle 216. The liquid reactant is thus dispersed from the nozzle outlet as a fog which absorbs radiant energy. The shroud 217 serves to assist in containing the liquid reactant fog centrally within a pre-reaction zone 411 of the reactor tube 401.

As shown best in Figures 6A and 7, the inlet assembly of the preferred embodiment of the present reactor may further include a plurality of secondary inlet tubes 218a, 218b, and 218c which enable the introduction of additional liquid reactants. The means for introducing the secondary liquid reactant are structurally and functionally similar to the means for introducing the principal liquid reactant, previously described, and thus further embody secondary atomizing gas inlet tubes 219a, 219b, and 219c and fogging nozzles such as 220a (the additional fogging nozzles are not shown). A representative inlet for a secondary liquid reactant and a representative inlet for a secondary atomizing gas are designated by reference numerals 221 and 222, respectively.

The above discussion presumes that the reactants themselves either exhibit a relatively high radiation absorption coefficient (α) or can be converted into a fog which absorbs radiant energy. However, if such is not the case, a radiant energy absorptive target, such as previously described, must be introduced into the

reactor zone 404 coincident with at least one point along the path of the reactants.

Referring particularly to Figure 6A, a sweep gas assists in directing the liquid reactant fog toward the reaction zone 404. The sweep gas enters nozzle block 204 through sweep gas inlet fitting 225, passes through channel 227 and is directed axially of the reactor tube 401 toward the pre-reaction zone 411. As shown in Figures 6A and 7, a reaction viewpoint 226 provides an axial view into the reaction zone 404.

B. Electrode Assembly

Referring particularly to Figures 6B, 8, 9 and 10, the tubular electrode assembly pressure vessel section 301 has first and second flange portions 303 (shown in Figure 6A) and 304, respectively. Electrode assembly pressure vessel section 301 is secured at its first flange 303 to the second flange 203 of the inlet assembly pressure vessel section 201 in fluidtight relationship. A coolant channel 305 is defined between the electrode assembly pressure vessel section 301 and an electrode assembly cooling jacket 306. Coolant enters the channel 305 through inlet 307 and exits through outlet 308.

As shown best in Figures 6B and 8, copper bus bar electrodes 309a—309f are mounted on and extend through the second flange 304 of the tubular electrode assembly pressure vessel section 301. Although there are six such electrodes 309, as a matter of convenience only one is actually shown in detail in Figure 7B. Each copper bus bar electrode 309 includes a phenolic flange 310 and a ceramic insulator 311. Each such electrode 309 is cooled by a fluid, preferably ethylene glycol, which circulates in an internal channel 312, entering through inlet 313 and exiting through outlet 314. An electrical connection is illustrated at 315. A polytetrafluoroethylene seal 316 assists in preventing any leakage from inside the inert fluid plenum 406. Although, as illustrated in Figure 16, the electrical system employed in connection with the present reactor is of the 3-phase "Y" connection-type, other systems may be used where circumstances warrant.

Referring particularly to Figures 6B and 6C, each copper electrode 309 is secured by a tongue and groove connection to a first extremity of a rigid carbon electrode extension 317. The electrode extensions 317 project through but do not contact a first end section 412 of the heat shield 410 and are secured at a second extremity to an arcuate heating element support 318. As shown best in Figure 9, heating elements 302a—302c are secured at a first end to one of the arcuate heating element supports 318 and are spaced circumferentially about the

reactor tube 401 within the inert fluid plenum 406. The heating elements are secured at a second end to a 3-phase center connection ring 319 as shown in Figures 6C and 10. Preferably, each electrically resistive heating element 302 is made of a fabric of a fibrous refractory material such as graphite or another form of carbon. Heating element supports 318 and center connecting ring 319 may be made of an electrically-conductive, refractory material such as carbon.

C. Main Assembly

Referring to Figures 6B, 6C and 9, the tubular main assembly pressure vessel section 405 has first and second flange portions 414 and 415, respectively. Section 405 is secured at its first flange 414 in fluid-tight relationship to the second flange 304 of the electrode assembly pressure vessel section 301. A main assembly coolant channel 416 is defined between the main assembly pressure vessel section 405 and a main assembly cooling jacket 417. The channel 416 is further defined by a spiral baffle 418. Coolant enters the spiral channel 416 through inlet 419 and exits through outlet 420.

The reactor tube 401 includes three zones: the prereaction zone 411, the reaction zone 404, and a post-reaction zone 422. As previously stated, the reactor tube 401 is made of a fabric of fibrous refractory material such as graphite or another form of carbon. The fabric may be knitted, woven, or nonwoven. The reaction tube 401 is secured at its outlet end 403 to a reactor tube outlet support ring 424 which, in turn, is secured in place by a reactor tube anchor block 425. The reactor tube 401 is secured at its inlet end 402 to a reactor tube inlet support ring 426 which, in turn, is joined in fluid-tight relationship to a tubular bellows 427 disposed within the pressure vessel inlet assembly section 201. An inlet end of the bellows 427 is secured in a fluidtight manner between the first flange 202 of the pressure vessel inlet assembly section 201 and the annular sealing flange 205 to insure that the inlet end of the reactor tube 401 remains sealed from the plenum 406. The bellows 427 is deformable to accommodate axial expansion and contraction of the reactor tube 401.

Means for applying an axial tensile force to the reactor tube 401 comprises three identical assemblies spaced equidistant about the circumferential surface of the pressure vessel inlet assembly section 201. For convenience, the assembly 428 which is illustrated in Figure 6A will be described. Each assembly 428 includes a translatable push rod 429 secured at one end to the reactor tube inlet support ring 426 and at an

opposite end to an annular plate 430. Each push rod 429 is supported in a bearing 431 which is sealed in a fluid-tight manner by O-ring 432. Eye-bolt 433 which is secured to the annular plate 430 anchors a cable 434 which extends generally parallel to the longitudinal axis of the reactor and over a pulley assembly 435. A weight 436 secured to an opposite end of the cable 434 applies a force which maintains the reactor tube 401 in axial tension.

Referring particularly to Figures 6B, 7B and 6C, the heat shield 410 includes a first circumferential section 438 which is disposed within the pressure vessel main assembly section 405, radially outwardly of the heating elements 302a, 302b and 302c and between the first end section 412 and a second end section 439 of the heat shield 410. As shown in Figure 6C, the first circumferential section 438 of heat shield 410 rests in a seating ring 437 which is preferably made of carbon. If desired, the first circumferential portion of the heat shield 410 may be extended in a direction toward the electrode assembly 300 to include a second circumferential portion 440 as shown in Figure 6B. Although molybdenum was the initial choice and had been found to be a satisfactory material for a heat shield of the type required in the present high temperature chemical reactor, it is preferred that the heat shield 410 of the present embodiment be made of a graphitic material such as pyrolytic graphite or a material manufactured by Union Carbide Corporation and sold under the trade name "Grafoil".

Radiometer viewports 441 and 442 are provided in the main assembly section 400. Viewport 442 enables observation and measurement of the temperature of the reaction zone 404 of the reactor tube 401 and viewport 441 enables observation and measurement of the temperature of heating element 302c.

D. Post-Reaction Treatment Assembly

As shown in Figure 6C, a first flange portion 502 of the post-reaction treatment assembly pressure vessel section 501 is secured in a fluidtight manner to a fluid-cooled interface flange 503 which, in turn, is secured in a fluid-tight manner to the second pressure vessel main assembly section flange 415. A coolant channel 504 is defined between post-reaction treatment assembly cooling jacket 505 and the post-reaction treatment assembly pressure vessel section 501. Coolant flows into the channel 504 through inlet 506 and exits through outlet 507. Radiometer viewport 509 is provided to enable observation and temperature measurement within the post-reaction zone 422 of the reactor tube 401.

Reaction products exiting the outlet end 403 of the reactor tube 401 of the embodiment of Figure 6 pass into a first section 510 of heat sink 511. As shown in Figure 6C and 6D, the first section 510 of the heat sink 511 includes an inner tubular wall 512 and an outer tubular wall 513 which define therebetween a coolant channel 514. Spiral coolant baffle 515 directs the coolant which enters through inlet 516 and exits through outlet 517. A first thermocouple probe 518 which extends into the first section 510 of the heat sink 511 enables the measurement of temperature of the entering reaction products. A second thermocouple probe 519 which extends into the first section 510 of the heat sink 511 measures the temperature of the reaction products about to exit.

Referring particularly to Figure 6D, the first section 510 of the heat sink 511 is joined to a second section 520 by flanges 521 and 522. The second section 520 includes an inner wall 524 and an outer wall 525 which define therebetween a coolant channel 526. Coolant enters the channel 526 through inlet 527 and exits through outlet 528. Thermocouple probes 530 and 531 enable measurement of the temperature of reaction products entering the second section 520 and exiting the second section 520, respectively.

Referring now to Figure 12 a post-reaction treatment assembly 500a suitable for use in the embodiment just described includes a post-reaction treatment assembly pressure vessel section 501a having a flange portion 502a which is secured in a fluid-tight manner to a fluid-cooled, interface flange such as flange 503 illustrated in Figure 6C. A coolant channel 504a is defined between a post-reaction treatment assembly cooling jacket 505a and the post-reaction treatment assembly pressure vessel section 501a. Coolant flows into the channel 504a through inlet 506a and exits through outlet 507a. Radiometer viewport 509a enables observation and temperature measurement in the post-reaction zone 422 of the reactor tube 401.

Reaction products exiting the outlet end 403 of the reactor tube 401 shown in Figure 12 at high temperature pass into a variable profile, counter-flow heat exchanger 532 which abuts the reactor outlet 403 at its inlet end 533. The heat exchanger 532 includes an inner tubular wall of refractory material 534, an outer tubular wall of refractory material 535 spaced concentrically outwardly from the inner wall 534, and a spiral baffle of refractory material 536 disposed between the walls 534 and 535 to define a spiral, annular coolant channel 537. The inner tubular wall 534,

outer tubular wall 535 and spiral baffle 536 together constitute a high temperature spiral heat exchanger assembly 544 which rests on a resilient carbon felt cushion 545 disposed on end plate 546 of heat exchanger pressure vessel section 547. Coolant inlets 538, 539 and 540 extend through the outer tubular wall 535 in communication with the spiral coolant channel 537.

In the specific post-reaction treatment assembly shown in Figure 12, after circulating throughout the spiral coolant channel 537 in a pre-selectable, variable, and controllable manner, the coolant is discharged at an outlet 541 of the spiral annular channel 537 adjacent the inlet end 533 of the heat exchanger 532. Thereafter, the coolant circulates through inlet port 542 in reactor tube anchor block 425a into the inert fluid plenum 406. In such case, it is apparent that the coolant employed should be a fluid which is the same as or, at least, compatible with the inert fluid which is present in the plenum 406. However, since the operation of the heat exchanger 532 does not require that the coolant be circulated into the plenum 406, alternative circulation patterns and expedients are feasible. In such instances, the choice of coolant fluid is not limited by the criteria set forth above. Circumferential heat exchanger cooling jacket 548 is spaced radially outwardly of the heat exchanger pressure vessel section 547, defining therebetween an annular channel 549. Coolant is introduced into channel 549 through inlet 550 and exits through outlet 551.

E. Inlet Assembly For Solid Reactants

The modified inlet assembly 200a shown in Figures 13A and 13B is substantially identical to the inlet assembly 200 of Figures 6A and 6B except that means for introducing a principal solid reactant of inlet assembly 200a replaces the means for introducing a principal liquid reactant of inlet assembly 200. For convenience, only the features of Figures 13A and 13B which differ from corresponding features of Figures 6A and 6B will be described.

A solid reactant inlet tube 232 extends through the annular nozzle block 204 and is fixedly secured thereto by a support flange 235. A principal solid reactant, preferably finely divided, enters inlet tube 232 through inlet 233 in support flange 235 and exists within reactor tube 401 adjacent the prereaction zone 411. Secured to and disposed radially outwardly of outlet 234 is a tubular shroud 217, the axis of which is substantially parallel to the axis of the reactor tube 401. Shroud 217 assists in containing finely divided solid reactants

centrally within the prereaction zone 411 of reactor tube 401.

Referring to Figure 14, a solid reactant feed system 238 is shown in combination with a high temperature reactor having an inlet assembly 200a of the type depicted in Figures 13A and 13B. A supply bin 240 for holding the solid reactant feeds a crusher 241, which, in turn, feeds a sieve 242. Coarse product output 245 of the sieve 242 is recycled to the crusher 241 and fine product output 243 is fed to a hopper 244 which is secured to an elongated tubular housing 246. Helical feed screw 247 is rotatably mounted within the housing 246 and is driven by motor 248. A pressure-sealing fluid may be introduced into the housing 246 through an inlet nozzle 249 located at a point downstream from the hopper 244; the interior of reactor tube 401 is thus sealed from the atmosphere. The solid reactant and the sealing fluid are discharged from housing 246 into the reactor through an outlet 250.

25 F. Refractory Coating And Etching Systems

For reasons set forth below, it is contemplated that a refractory coating may be deposited on surfaces of reactor tube 401, heating elements 302, and heat shield 410 which are exposed to the blanket gas and to high temperature during operation of the reactor. Such refractory coating may be, for example, pyrolytic carbon or a refractory oxide such as thorium oxide, magnesium oxide, zinc oxide, aluminium oxide, or zirconium oxide. It is further contemplated that portions of the surface of the reactor tube 401 may be selectively etched or eroded.

Referring to Figure 15, a refractory coating and etching system 600 is schematically represented and comprises a first refractory deposition agent metering system 601 having a carbonaceous gas supply 602 connected to a carbonaceous gas metering line 603. The metering line 603 has an on/off valve 604 connected to a needle valve 605 and a flow meter 606. A first feeder line 608 connects the carbonaceous gas metering line 603 to an admixture gas supply line 607.

A second refractory deposition agent metering system 610 includes a carrier gas supply 611 connected to a carrier gas metering line 612 which has an on/off valve 613, a needle valve 614, and a flow meter 615. The carrier gas metering line 612 is connected to a bubble tube 616 disposed within a tank 617 which contains a solution of a volatile metal-containing compound. The temperature of the tank 617 is regulated by a temperature controller 618 which senses the temperature of the tank by

a thermocouple 619 and supplies heat to the tank, as required, by an electric heating mantle 620. An outlet end 621 of bubble tube 616 is submerged in the solution contained in the tank 617. An outlet 622 of the tank 617 connects a second feeder line 623 to the tank 617 at a point above the solution surface. The second feeder line 623 is also connected to the admixture gas supply line 607.

In an etching agent metering system 625, an etching agent supply 626 is connected to an etching agent metering line 627 which includes, in series, an on/off valve 628, a needle valve 629, and a flow meter 630. Connected to the etching agent metering line 627 is a third feeder line 631, which is connected to the admixture gas supply line 607.

The three lines 608, 623, and 631, all feed into the admixture gas supply line 607, which branches at a T-joint 632. A first branch line 633 includes a first branch line valve 634 and is connected to a first inlet of an inert fluid mixing manifold 635. A second branch line 636 includes a second branch line valve 637 and is connected to a first inlet of a sweep gas mixing manifold 638.

An inert fluid supply 640 is connected to an inert fluid metering line 641 which includes an on/off valve 642, a needle valve 643 and a flow meter 644 which is connected to a second inlet of inert fluid mixing manifold 635. An outlet of mixing manifold 635 is connected to an inert fluid supply line 645 which, in turn, is connected to the pressure vessel inlets 408 and 409 for directing the inert fluid into the inert fluid plenum 406. A plenum pressure sensor 646 is connected to the inert fluid supply line 645 and is in communication with the plenum 406 for measuring the pressure of the inert fluid within the plenum. A plenum exhaust valve 647 is also connected to the inert fluid supply line 645 and provides an outlet for discharging fluid from the plenum.

A sweep gas supply 648 is connected to a metering line 649 which includes an on/off supply valve 650, a needle valve 651, and a flow meter 652 which is connected to a second inlet of the sweep gas mixing manifold 638. An outlet of mixing manifold 638 is connected to a sweep gas supply line 653 which, in turn, is connected to the sweep gas inlet fitting 225 for introducing the sweep gas into the interior of the reaction tube 401. A reaction zone pressure sensor 654 which connects to the sweep gas supply line 653 and which communicates with the interior of the reactor tube 401, measures the pressure in the reaction zone of the reactor.

As shown best in Figure 6D, a reactor

tube outlet closure valve 655 is secured to the second section 520 of the heat sink 511 by flanges 555 and 656.

5 When the reactor is in operation, a pressure differential must be maintained between the inert fluid in plenum 406 and gas in the reactor tube 401 to cause a uniform flow of inert fluid radially inward through the porous wall of the tube 401. It is thus advantageous that the fabric of tube 401 be sufficiently stiff that the pressure differential may be maintained without inward collapse of the tube 401. Accordingly, it is contemplated that a refractory coating such as pyrolytic carbon be deposited upon portions of the fibrous refractory material of the reactor tube 401 which are disposed within the heat shield to increase the stiffness or dimensional stability of the fabric.

20 To deposit such coating, reactor tube outlet closure valve 655 is closed and the reactor tube 401 is heated to about 3450°F. Next, the on/off valve 650 in the sweep gas metering line 649 is opened, the on/off valve 642 in the inert fluid metering line 641 is closed, and the plenum exhaust valve 647 is opened, permitting sweep gas to flow into the interior of the reactor tube 401, then radially outwardly through the porous wall of the tube 401 into the plenum 406, and, finally through the pressure vessel inlets 408 and 409 and the plenum exhaust valve 647. This tends to expand the tube 401 to its maximum diameter. Thereafter, the on/off valve 604 in the carbonaceous gas metering line 603 is opened. The needle valves 605 and 651 are adjusted to set the flow rates of the carbonaceous gas and the sweep gas, respectively, to suitable values as registered on flow meters 606 and 652. The first branch line valve 634 is closed and the second branch line valve 637 is opened so that the carbonaceous gas flows through the first feeder line 608, the admixture gas supply line 607, the T-joint 632, the second branch line 636, and into the sweep gas mixing manifold 638 where it mixes with the sweep gas and flows into the interior of the reactor tube 401 through sweep gas supply line 653 and sweep gas inlet fitting 225.

50 The carbonaceous gas dissociates on the heated surfaces which it contacts, depositing a pyrolytic graphite coating. Thus, pyrolytic graphite is generally deposited on the portions of the reactor tube 401 and the heating elements 302 within the heat shield 410 and on the interior of the heat shield 410.

60 Since the portion of the reactor tube 401 which lies within the pre-reaction zone 411 is outside the heat shield 410 and, thus, may not be heated conveniently to temperatures above the decomposition temperature of the carbonaceous gas, it is contemplated

that a stainless steel screen 450, shown in Figures 6A and 6B, be provided to prevent the flexible reactor tube 401 from collapsing inwardly under the pressure differential of the inert fluid, although it has been found that increased tension on the porous fabric accomplishes substantially the same result.

To control the rate of flow of inert fluid through the walls of the reactor tube 401, the diameter of the pores in the tube wall may be reduced or enlarged while the reactor is in operation by mixing a refractory deposition agent or an etching agent with the inert fluid. The pressure differential between the plenum and the reaction zone may be monitored by the pressure sensors 646 and 654 and the rate of flow of inert fluid through the wall may be monitored by the flow meter 644.

When the pressure differential becomes too low at the desired rate of flow of inert blanking gas, the diameter of the pores in the tube of the reactor wall may be reduced by opening the on/off valve 604 and adjusting the needle valve 605 to allow a carbonaceous gas from the carbonaceous gas supply 602 to flow through carbonaceous gas metering line 603. The second branch line valve 637 is closed and the first branch line valve 634 is opened to direct the carbonaceous gas into the inert fluid mixing manifold 635 and thence into the plenum 406 through the inert fluid supply line 645 and the pressure vessel inlets 408 and 409. The plenum exhaust valve 647 remains closed and the reactor tube outlet closure valve 655 remains open during normal operation of the reactor. The carbonaceous gas dissociates on the heated surfaces within the reactor which it contacts. Accordingly, carbonaceous gas which flows into the pores of the fabric of the wall of the reactor tube 401 dissociates, depositing a coating of pyrolytic graphite which reduces pore diameter. Since the pressure differential across the reactor tube wall will increase for a fixed flow of inert fluid, the decrease in porosity of the tube may be monitored with pressure sensors 654 and 646 and flow meter 644 as the graphite is deposited. When the pressure differential exceeds a predetermined value, the growth of the graphite coating may be halted by closing the on/off valve 604 in the carbonaceous gas metering line 603. The entire process of reducing the diameter of the pores in the reactor tube wall may be carried out without interrupting the operation of the reactor.

Conversely, it may be necessary to increase the diameter of the pores of the reactor tube 401. In this case, an etching agent such as steam or molecular oxygen

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from the etching agent supply 626 is mixed with the inert fluid by opening valve 628, adjusting needle valve 629 in the etching agent metering line 627, closing the second branch line valve 637, and opening the first branch line valve 634. The etching agent mixes with the inert fluid in inert fluid mixing manifold 635 and flows into the plenum 406 through the pressure vessel inlets 408 and 409. The etching agent attacks heated surfaces which it contacts, thereby increasing the diameter of the pores of the heated portion of the reactor tube 401. The flow of etching agent may be continued until pressure sensors 654 and 646 indicate a sufficiently low pressure differential across the reactor tube 401 for the desired rate of flow of inert fluid as monitored by flow meter 644. As with reducing the pore diameter with the carbonaceous gas, this process may be carried out while the reactor is in operation.

It may be advantageous in some applications to use steam or another corrosive medium as the blanket inert fluid. To prevent or, at least, to retard the corrosion of materials of which the reactor is constructed, it is contemplated that a coating of a refractory oxide such as thorium oxide, magnesium oxide, zinc oxide, aluminium oxide, or zirconium oxide be deposited on the portions of the reactor tube 401, heating elements 302, and heat shield 410 which come into contact with the inert fluid and operate at high temperatures. To deposit a coating of refractory oxide, a refractory deposition agent which is a volatile metal-containing compound such as methylmagnesium chloride, magnesium ethoxide, or zirconium-n-amyloxide may be employed. Methylmagnesium chloride, for example, decomposes on a surface heated to about 1100°F. to deposit a coating of magnesium metal. The hot magnesium metal is subsequently oxidized by introducing steam or molecular oxygen into the plenum 406. Zirconium-n-amyloxide and magnesium ethoxide both generally decompose on heated surfaces to form zirconium oxide or magnesium oxide respectively.

Referring to Figure 15, the volatile metal-containing compound may be introduced into the plenum 406 by causing a carrier gas from the supply 611 to flow through the metering line 612 by opening the on/off valve 613. The needle valve 614, adjusts carrier gas flow rate to a suitable value as measured by flow meter 615. The tank 617 contains, for example, a solution of the volatile metal containing compound such as methylmagnesium chloride dissolved in diethyl ether or zirconium-n-amyloxide dissolved in tetrahydrofuran.

The carrier gas flows through the bubble tube 616 and into the solution of tank 617. The second branch line valve 637 remains closed and the first branch line valve 634 remains open in order that the carrier gas, solvent vapor, and metal-containing compound vapor are directed sequentially through the outlet 622 of the tank 617, the second feeder line 623, the admixture gas supply line 607, and the first branch line 633, and into the inert fluid mixing manifold 635 where they are mixed with the inert fluid and then carried to the plenum 406 over the inert fluid supply line 645 and through the pressure vessel inlets 408 and 409. The volatile, metal-containing compound decomposes on hot surfaces which it contacts within the reactor. If it decomposes into a pure metal, oxygen or steam is subsequently introduced into the plenum 406 to cause formation of the oxide.

G. Process Variable Control Systems

Figure 16 illustrates a temperature control system 700 for the reactor of Figures 6A to 12 (the second embodiment). In Figure 16, heating elements 302a, 302b and 302c are depicted in schematic form connected in a "Y" configuration circuit, one end of each heating element being connected to a tie point 701 and the other end being connected to a branch 702a, 702b, or 702c of a three-phase power line 702. The tie point 701 corresponds to the three-phase connecting ring 319 of Figure 6C. The power line 702 connects to a heater power output 703 of a power controller 704, which, in turn, connects to a principal threephase power line 705 and a firing circuit 706. The principal three-phase power line 705 supplies current, preferably at 440 volts, for heating the reactor. A radiometer 708 disposed within the viewport 441 of Figure 6B is focussed on the heating element 302c and produces a signal, generally in the millivolt range, which corresponds to the temperature of the heating element. An "MV/I" converter 709 amplifies the radiometer signal and converts it to an electric current. A setpoint controller 707, an output signal line 712 for connection to a computer (not shown), and a recorder 710 which makes a permanent log of the temperature measured by the radiometer 708 are all connected to the converter 709. An input signal line 713 connects a control signal input 711 of the setpoint controller 707 to a computer (not shown). Current meters 750a, 750b, and 750c are inserted in the three branches 702a, 702b, and 702c, respectively, to measure the current supplied to heating elements 302a—c; and voltmeters 751a, 751b, and 751c are tied to the branches 702a—c to measure the voltages across the

heating elements. The power dissipated in the heating elements and the electrical resistance of the heating elements can be calculated from such voltage and current measurements. Knowledge of the electrical resistance of each heating element provides information as to its physical integrity since, as a heating element erodes, its electrical resistance increases.

Figure 17 is a graph of the electrical sheet resistance of a sample of graphite cloth (sold under the trade name of "WCA Graphite Cloth" by Union Carbide Corporation) as a function of the temperature of the cloth. The cloth has been stiffened with pyrolytic graphite by heating and exposing it to an atmosphere of a carbonaceous gas, generally according to the procedure described above. The vertical axis of the Figure 17 graph gives the sheet resistance in units of "ohms per square" since, as is known, the resistance measured between opposing edges of squares of a resistive material of a given thickness is independent of the dimensions of the square. Thus, the resistance at a particular temperature of a heating element formed from a single rectangular strip of "WCA Graphite Cloth" may be found by considering the strip to be made up of squares of the cloth connected in series. For example, the resistance of a strip 6 inches by 51 inches at 2500°F. measured between the two six-inch sides is found by multiplying (51/6) times 0.123 ohms, the sheet resistance at 2500°F. given on Figure 17. The resistance of a heating element made up of more than one layer of fabric, each layer having the same dimensions and therefore the same resistance, is found by dividing the resistance of a single layer by the number of layers. For convenience, the calculated sheet resistances in "ohms per square" for samples of stiffened "WCA Graphite Cloth" made up of 2, 3, and 4 layers have also been plotted on Figure 17.

In operation, after the setpoint controller 707 is set to a specified temperature either manually or by a computer, it compares such temperature with the measured temperature of the electrode 302c and produces an error signal which depends upon the algebraic difference between the measured temperature and the specified temperature. The setpoint controller 707 controls the firing circuit 706, which, in response to the error signal, causes the power controller 704 to increase or decrease the power supplied to the heating elements to reduce, as necessary, the magnitude of the error signal, causing the temperature of the heating element 302c to approach the specified temperature. Because the heating element 302c is within the zone enclosed by the heat shield 410, its

temperature is generally representative of the temperature of surfaces throughout the enclosed zone. However, radiometers focussed on other surfaces within that zone may also be used for temperature control.

As shown in Figure 18, process variables in addition to temperature may be regulated by feedback control systems as, for example, a principal liquid reactant feed rate regulation system 714 which includes a supply 715 communicating with a metering system 716 over a feed line 717. The metering system 716 controls the flow rate of the principal reactant and may include, for example, a variable speed pump and pump controller or a variable orifice valve and valve controller. An output 718 of the principal reactant metering system 716 is connected to a flow rate transducer 719 which produces an electrical signal output 720 corresponding to the rate of flow of the principal reactant. An output 721 of the principal reactant flow rate transducer 719 is connected to the principal liquid reactant inlet pipe 215. A signal output 722 of the reaction zone pressure sensor 654 and the signal output 720 of the flow rate transducer 719 are connected to the first and second signal inputs, respectively, of the principal reactant metering system 716. An output of a computer system 723 is connected to a third input of the metering system 716.

In one mode of operation of the principal liquid reactant feed rate regulation system 714, the computer system 723 communicates both a pre-selected value for the principal reactant flow rate and an upper limit for the reaction zone pressure to the principal reactant metering system 716 which compares the pre-selected flow rate with that measured by the transducer 719 and adjusts the flow rate to approach the selected value, provided, however, that the reaction zone pressure is below the prescribed upper limit. Should the reaction zone pressure exceed this upper limit, the metering system 716 will lower the pressure by reducing the flow rate of the principal reactant.

A secondary liquid reactant flow rate regulation system 724 is another feedback control system which includes a supply 725 communicating with a metering system 726 over a feed line 727. The secondary reactant metering system 726 may be of the same type as the principal reactant metering system 716. An output 728 of the secondary reactant metering system 726 is connected to a flow rate transducer 729 which produces a signal corresponding to the rate of flow of the secondary reactant. An output 731 of the transducer 729 is connected to the secondary reactant inlet

221. A signal output 722 of the reaction zone pressure sensor 654 and a signal output 730 of the secondary reactant flow rate transducer 729 are connected to separate signal inputs of the secondary reactant metering system 726, and an output of the computer system 723 is connected to a third input. The secondary liquid reactant flow rate regulation system 724 may be operated in a mode analogous to that described above for the principal liquid reactant regulation system 714.

In an inert fluid flow rate regulation system 734, an output of the inert fluid supply 640 is connected to the needle valve 643, which, in turn, is connected to the on/off valve 642. Valve 642 is connected to an inert fluid flow rate transducer 735. A signal output 736 of the transducer 735 is connected to a first input of an inert fluid needle valve controller 737. A second input of the needle valve controller 737 is connected to the computer system 723 and a third input is connected to the plenum pressure sensor 646. The opening of the needle valve 643 may be set by the controller 737. An inert fluid output of transducer 735 is connected to the pressure vessel inlets 408 and 409 of the reactor. For convenience, the plenum exhaust valve 647, flow meter 644 and inert fluid mixing manifold 635, of Figure 15 are not shown in Figure 18, and the inert fluid flow rate transducer 735 of Figure 18 is not shown in Figure 15.

In operation, the on/off valve 642 is opened, allowing the inert fluid to flow through transducer 735 and into the inlets 408 and 409. The needle valve controller 737 compares a flow-rate signal from the transducer 735, to a flow rate specified by the computer system 723 and adjusts needle valve 643 accordingly, provided, however, that the plenum pressure as sensed by pressure sensor 646 does not exceed an upper limit also specified by the computer system 723. If the pressure is excessive, the needle valve controller 737 reduces the flow rate to lower the pressure.

A reactor temperature control system 700, shown in detail in Figure 16 and depicted schematically in Figure 18, comprises a reactor temperature controller 738 which includes the power controller 704, firing circuit 706, set point controller 707, converter 709, recorder 710, and meters 750 and 751 shown in Figure 16. The radiometer 708 (not shown in Figure 18) is housed within the viewport 441 and connected to the controller 738. The three-phase power line 702 connects the heater power output 703 of the reactor temperature controller 738 to the heating elements 302 (not shown in Figure 19) through the electrodes 309. Thus, the level

of electrical power supplied at the heater power output 703 determines the temperature of the reactor tube 401. The control signal input 711 and an output of the reactor temperature controller 738 are connected to the computer system 723 by the input signal line 713 and the output signal line 712, respectively.

A reactor product sampler 740, connected to an outlet 741 located adjacent the reactor outlet closure valve 655, transfers at preselected time intervals samples of reaction product into a sample inlet 742 of a gas chromatograph 743. An electrical signal at an output 744 of the chromatograph 743 responds to changes in the chemical composition of the samples. For example, the gas chromatograph 743 in conjunction with the reaction product sampler 740 may produce a signal which corresponds to the concentration of ethylene in a process for the partial pyrolysis of a hydrocarbon.

Outputs of the gas chromatograph 743 are connected to a recorder 749 and the computer system 723. An input 745 of the computer system 723 is connected to transducers for the process variables by a data bus 746, which includes signal lines connected to the flow rate transducer 719, 729 and 735, pressure sensors 646 and 654, temperature controller 738, and gas chromatograph 743. Other transducers may be tied to the data bus 746 as desired. An output 747 of the computer system 723 is connected to a command bus 748 which includes signal lines tied to the principal reactant metering system 716, secondary reactant metering system 726, reactor temperature controller 738, and inert fluid needle valve controller 737. The computer system 723 may include a digital computer, an analog-to-digital converter for converting analog signals of the transducers to digital data for the computer, a digital-to-analog converter for converting digital signals from the computer to analog control signals, and a multiplexer for switching among signal lines in the data bus 746 and the command bus 748.

It is contemplated that during a process run the computer system 723 may specify and monitor process variables by signals communicated over the command bus 748 and the data bus 746. Thus, the computer system 723 may supervise the operation of the reactor to ensure that process variables remain within specified ranges. Moreover, the computer may be programmed to search for optimum operating conditions for a particular process by making systematic variations in the process variables while monitoring the output of the reactor with the chromatograph 743. For example, the computer may be

programmed to search for reactor temperatures and feedstock flow rates which maximize the ethylene concentration in the output for a particular hydrocarbon feedstock. The computer system 723 may also be incorporated in feedback control systems; such as a reaction product control system which includes in addition to the computer system 723 the reaction product sampler 740, the gas chromatograph 743, the reactor temperature controller 738 which includes an SCR circuit, and the three-phase power line 702 connected to the heating elements 302 through the SCR circuit. In this reaction product control system, the computer system compares the chemical composition of samples of reaction product withdrawn from the reactor to a preselected composition and generates an electrical signal at its output 747 corresponding to deviations in the chemical compositions of the samples. The output 747 of the computer system 723 is connected to the input 711 of the reactor temperature controller to enable variation of the temperature of the reactor tube in response to changes in the signal from the computer system, reducing the deviations in the chemical composition of the reaction products. Other process variables such as the feedrates of selected reactants and the pressure in the reaction zone may also be controlled by similar feedback control systems.

High temperature chemical reaction processes conducted in accordance with the present invention necessitate the use of a fluid wall substantially transparent to the radiation employed, the wall preferably being in the form of an annular envelope or blanket of the inert fluid having a substantial axial length. The annular envelope is preferably generated in a direction generally perpendicular to its axis and radially inwardly of its outer circumferential surface. The integrity of the fluid envelope is thus independent of any flow considerations and may be maintained for an axial distance much greater than that obtainable if the envelope were generated in a direction generally parallel to its axis. The primary requirement is to maintain the flow of the inert fluid under a greater pressure than that of the reactant stream to prevent the reactants from "punching through" or otherwise breaking out of confinement within the envelope.

After the envelope has been generated, the process of the invention is preferably conducted as follows. At least one reactant is passed through the core of the envelope along a predetermined path which is substantially coincident with the envelope axis. The envelope confines the reactants

therewithin and out of contact with the containing surfaces of the reactor chamber.

Finally, high intensity radiant energy is directed into the envelope core to coincide with at least a portion of the predetermined path of the reactants. Such radiant energy may be directed to at least one point along the path of the reactants or, preferably, it may be directed along a finite length of the path as contemplated by the two embodiments described in detail above. In either case, sufficient radiant energy is absorbed in the core to raise the temperature of the reactants to a level required to sustain the desired chemical reaction.

In the event that the reactants will not themselves absorb radiant energy, an absorptive target may be introduced along the path of the reactants. The target will then absorb sufficient radiant energy to raise the temperature in the core to the level required to initiate the desired chemical reaction. As previously stated, if the contemplated reaction is such that the transparent reactants produce at least one product which absorbs radiant energy, the target may be withdrawn, or its supply discontinued after the reaction has been initiated.

The contemplated process may further include the step of cooling the reaction products and any remaining reactants and/or targets immediately after the desired reaction has been completed. The purpose of this procedure is to terminate the desired reaction and to prevent the occurrence of any further undesired reaction. The products, targets and remaining reactants may be cooled conveniently and effectively by radiation heat transfer to a cool, radiant energy absorbing surface.

The fluid-wall reactors of the invention may be used in virtually any high temperature chemical reaction, many of which reactions have been previously regarded as either impractical or only theoretically possible. The most important criterion for utilizing these fluid-wall reactors is a particular high thermodynamically possible under the reaction conditions. Utilizing these fluid-wall reactors, such high temperature chemical reaction processes can be conducted at temperatures up to about 6000°F.

Among the reactions which may be carried out in the fluid-wall reactors of the invention are the dissociation of hydrocarbons and hydrocarbonaceous materials, such as coal and various petroleum fractions, into hydrogen and carbon black; the steam reforming of coal, petroleum fractions, oil shale, tar sands, lignite, and any other carbonaceous or

hydrocarbonaceous feedstock into synthesis gas mixtures, which processes may also include the optional use of one or more inorganic carbonates (such as limestone or dolomite) or inorganic oxides to chemically react with any sulfur-containing contaminants such that they may be removed from the resultant synthesis gas mixtures; the partial dissociation of hydrocarbons and hydrocarbonaceous materials into lower molecular weight compounds; the partial pyrolysis of saturated hydrocarbons into unsaturated hydrocarbons, such as ethylene, propylene and acetylene; the conversion of organic waste materials, such as sewage sludge or lignin-containing by-products, into a fuel gas; the complete or partial desulfurization of sulfur-containing hydrocarbonaceous feedstocks; the reduction of mineral ores or inorganic compounds to the element or to lower-oxidation-state compounds with a reducing agent, for example, hydrogen, carbon, carbon monoxide, or synthesis gas (i.e. a gas mixture comprising carbon monoxide and hydrogen); and the partial or complete reaction of an inorganic element or compound with a carbonaceous or hydrocarbonaceous material to form the corresponding inorganic carbide.

If desired, one or more catalysts may be used in such high temperature chemical reaction processes to accelerate the reaction or to change its course to a desired reaction sequence. Where such processes involve carbonaceous or hydrocarbonaceous reactants, the addition of an appropriate catalyst to the system may be used to promote the formation of free radicals, carbonium ions or carbanions to influence the course of the reaction.

Of course, no one set of operating conditions is optimum or appropriate for all reactions which may be carried out in the fluid-wall reactor. Operating conditions, such as temperatures, pressures, rates of feed, residence time in the reactor tube, and rates of cooling, may be varied to match the requirements for the particular reaction conducted. By way of illustration, among the factors which influence the products of the pyrolysis of a hydrocarbon are the temperature to which the hydrocarbon is heated and the length of time it is maintained at that temperature. It is known, for example, that methane must be heated to about 2250°F. in order to produce acetylene. Ethylene formation from ethane begins at a lower temperature, about 1525°F. In a typical process for pyrolyzing hydrocarbons, acetylene, ethylene, hydrogen, carbon black, and hydrocarbon oils are produced. Reaction times on the order of a millisecond

generally maximize the yield of acetylene, since reaction times of greater than a millisecond generally favor the production of ethylene and other products at the expense of acetylene, while reaction times of less than a millisecond generally reduce the yields of both ethylene and acetylene. Very high temperatures, for example in excess of 3000°F., generally favor the production of carbon black and hydrogen at the expense of acetylene and ethylene. Reaction times in the fluid-wall reactors of the invention may be shortened by shortening the reactor tube and by increasing the rate of flow of reactants introduced into the reactor tube. For very short reaction times, it may be advantageous to mix a radiation-absorbing target, such as carbon black, with the reactants in order to promote efficient coupling between the reactant stream and the thermal radiation from the tube wall and thereby facilitate heating the reactants quickly.

EXAMPLES

The following Examples are illustrative of the ease with which various high temperature chemical reaction processes may be carried out in fluid-wall reactors in accordance with the invention. In each of these Examples, the high temperature fluid-wall reactor illustrated in Figures 1 to 5 and referred to as the first embodiment was utilized to carry out the particular high temperature reaction. The reactor tube 61 was a porous graphite tube 36 inches in length which had an inside diameter of 3 inches and an outside diameter of 4 inches, the average pore radius being 20 microns. The porous tube was encased in a steel pressure vessel 70, which was 10 inches in diameter. Reactor tube 61 was heated by carbon heating elements 100a to 100f, which were disposed within plenum 85. The heat shield 120, also located within plenum 85, was made of molybdenum. A water-cooled collar 125 was located adjacent to the outlet end of reactor tube 61 to cool the reaction products formed by radiation coupling. After each Example had run continuously for various periods of time, the reactor tube 61 was inspected for buildup of carbon back or other material. None was found.

EXAMPLE I

Thermal Dissociation of Methane

A series of tests was conducted to determine the effectiveness of the fluidwall reactor in thermally dissociating natural gas at various feedrates and reaction temperatures. In each of these tests, hydrogen was introduced into plenum 85 through inlet 83 and forced through porous

reactor tube 61 into the reactor chamber at a constant rate of 5 scfm. The current through carbon electrodes 100a—100f was adjusted to set the temperature of the reactor tube from 2300° to 3400°F., as measured with an optical pyrometer. Natural gas, consisting of greater than 95% methane with the balance being ethane and propane, was introduced into the reactor through inlet 91 at various flow-rates ranging from 1 to 5 scfm. A small amount of carbon black was introduced into the reactor at the same time through inlet 121 to serve as an absorbent target for the purpose of initiating the pyrolytic dissociation. Once the dissociation had begun, it was not necessary to add additional carbon black to sustain the reaction. A dense black smoke streamed from the outlet end of the reactor tube and was found to consist of carbon black and hydrogen. The carbon black particles were extremely fine and difficult to filter. By spraying water into the effluent stream just below the outlet end of the reactor tube 61, it was possible to agglomerate the carbon black particles and collect them on a cloth dust filter. Table I sets forth the percent dissociation at various flow-rates ranging from 1 to 5 scfm and at dissociation temperatures ranging from 2300° to 3400°F., the fraction of methane dissociated being determined by measuring the thermal conductivity of the effluent gas after filtering the carbon black particles from the sample.

TABLE I
Percent Dissociation At Various Flow-Rates And Temperatures

Dissociation Temperature (°F.)	Flow-Rate (scfm)				
	1	2	3	4	5
2300°	86	74	66	60	54
2500°	89	79	72	68	63
2700°	91.5	83	78	74.5	70.5
2900°	94	88	84.5	82.0	79
3000°	95.5	91	88.5	86	83.5
3100°	97	94	92.5	91.0	89.5
3200°	98.5	98.5	98.5	98.5	98.5
3300°	100	100	100	100	100
3400°	100	100	100	100	100

EXAMPLE II Thermal Dissociation Of Liquid Hydrocarbons

A series of tests were performed to determine the effectiveness of the fluid-wall reactor in thermally dissociating liquid hydrocarbons. Hydrogen was used as the blanket gas at constant flow rate of 5 scfm. The liquid hydrocarbons selected for the test series were typical distillates obtained from crude petroleum and included naphtha (b.p. 100° to 200° F.); kerosene-diesel (b.p. 220° to 350° F.); gas oil (b.p. 350° to 600° F.); and residual oil and asphalt (b.p. > 600° F.). The results of these tests were as follows:

A. Naphtha. A stream of naphtha at approximately 80°F. was fed into reactor tube 61 at a rate of 0.05 gallon per minute through inlet 121. The temperature of the reactor tube was held at 3400°F. The pure naphtha passed through the reactor unaffected, apparently being transparent to the thermal radiation emanating from the incandescent reactor tube. The naphtha was then made opaque by mixing it with 0.1% by weight of finely divided carbon black. When this opaque mixture was introduced into the reactor as before, there was an

excellent coupling with the thermal radiation. Carbon black and hydrogen streamed from the outlet of the reactor tube. An analysis of the product gas with a thermal conductivity cell showed it to be greater than 98 mole % hydrogen, indicating that the dissociation was nearly complete.

B. Kerosene-Diesel. Kerosene-diesel was mixed with 0.1% by weight carbon black and then fed into the fluid-wall reactor at a rate of 0.05 gallon per minute. The reactor tube was held at 3400°F. The kerosene-diesel dissociated into carbon black and hydrogen. Thermal conductivity measurements indicated that the effluent gas consisted of greater than 98 mole % hydrogen.

C. Gas Oil. Gas oil mixed with carbon black was introduced into the fluid-wall reactor at a flow rate of 0.05 gallon per minute. When the reactor tube was held at 3400°F., the gas oil dissociated into carbon black and hydrogen, which, when separated from the carbon black, was found to consist of 98 mole % pure hydrogen, based on thermal conductivity measurements. When the temperature of the reactor tube was decreased to 2800°F., the effluent from the reactor changed from a dense black

smoke to a light gray fog, indicating that at the lower reaction temperature the gas oil was only partially dissociated, probably into lighter hydrocarbon fractions and a small amount of carbon.

5 D. Residual Oil And Asphalt. Residual oil containing asphalt, introduced into the fluid-wall reactor at 0.05 gallon per minute, completely
10 dissociated into carbon black and hydrogen when the reactor tube was held at 3400°F. Thermal conductivity analysis of the gaseous component of the effluent stream showed that it was greater than 98 mole %
15 hydrogen.

EXAMPLE III

Thermal Dissociation Of Coal

A sample of Utah soft coal was analyzed and found to contain 0.58% by weight of sulfur and 8.55% by weight of ash. The coal
20 was pulverized to -50 mesh and fed into the reactor at approximately 35 pounds per hour. Reactor tube 61 was held at 3000°F. and was protected by a blanket of nitrogen, which was forced through the porous wall
25 at a rate of 5 scfm. The coal dissociated into carbon black, gaseous products, and a light coke.

The carbon black differed from that produced in Example I in that the particles
30 were sufficiently large to filter without the addition of water. The carbon black was found to contain 8.63 weight % ash and 0.54 weight % sulfur. The gaseous product was a mixture of hydrogen and nitrogen (the
35 latter from the blanket gas), containing only 0.02 mole % sulfur, which was present as hydrogen sulfide.

Approximately 62% by weight of the starting material was converted into coke. This coke was extremely light and open; its
40 density was only 35% of the density of the coal from which such coke was made. When freshly prepared, the coke spontaneously oxidized in air to an ash in less than 12 hours, indicating that it had
45 high surface activity. When the coke was allowed to remain at room temperature in a nitrogen atmosphere overnight, it did not show evidence of surface activity and did
50 not spontaneously oxidize when subsequently exposed to air. Microscopic examination of the coke showed that it consisted of small, hollow, spherical globules of a glasslike substance. Chemical
55 analysis showed that the coke contained 8.27 weight % ash and 0.70 weight % sulfur.

EXAMPLE IV

Steam Reforming And Gasification Of Coal

A sample of coal from Carbon County, Utah, which contained an ash with a high limestone content, as analyzed and found to contain 0.60% by weight of sulfur. The coal was pulverized to -50 mesh and fed
60 into the reactor at approximately 10.45 pounds per hour. Steam at a temperature of 250°F. was simultaneously introduced into the reactor at a rate of 20 pounds per hour. Reactor tube 61 was held at 3400°F. and
65 was protected by a blanket of hydrogen which was forced through the porous wall at a rate of 5 scfm. A dense white vapor was observed to emanate from the outlet of the reactor. There was no evidence of any
70 carbon black or heavy residue having been produced. No ash or other solid material was found in the hopper located directly beneath the reactor tube outlet, indicating that all of the solid residue in the coal was
75 entrained in the gaseous product.

The solid products were filtered from the effluent stream and the remaining gas was dried prior to analysis with a mass spectrometer. The results of the analysis, neglecting air, are as follows
85 (concentrations being given in mole percent): nitrogen (0.051%); carbon monoxide (7.563%); hydrogen sulfide (none observed); carbon disulfide (none observed); carbon dioxide (0.277%);
90 hydrogen (89.320%); methane (1.537%); other hydrocarbons, such as benzene, acetylene, etc. (1.253%).

The gaseous product from this reaction is suitable as a fuel. Moreover, no sulfur-containing components were observed in the analysis, although the mass spectrometer was capable of detecting sulfur
95 compounds in concentrations as low as 10 ppm. This indicated that essentially all of the sulfur initially present in the coal had been entrained in the solid particles which were filtered from the effluent stream.

EXAMPLE V

Steam Reforming And Gasification Of Oil Shale

A sample of Green River oil shale, obtained from a source near Ruffle, Colorado, was pulverized to a -100 mesh
100 size. The sample was analyzed for the various carbonaceous materials present in oil shale. Methylene chloride at room temperature extracted 0.93 weight % of the

shale. The sample was further analyzed by heating a portion of it in air and observing the weight loss as a function of temperature. The results of such further analysis were as follows:

	Temperature Range	Weight Loss %	Remarks
5	68°—932°F.	11.60	distillation of volatiles
	932°—1436°F.	2.50	oxidation of carbon
	1436°—2192°F.	12.00	decarboxylation of CaCO ₃

From these measurements it was estimated that the oil shale was composed of 15 weight % of organic material and 27.3 weight % of limestone as CaCO₃. The remaining 57.7% by weight was assumed to be siliceous material.

The pulverized sample was introduced into the reactor at a rate of 38 pounds per hour. Simultaneously, steam was fed into the reactor at approximately 20 pounds per hour. The steam was at a temperature of 250°F. at the inlet to the reactor. The tube was maintained at a temperature of 3100°F., and hydrogen, injected through the porous wall at a rate of 5 scfm, served as the blanket gas. A water-white vapor streamed from the outlet end of the tube. The temperature of this vapor stream was measured to be 970°F. just below the outlet of the reactor.

A solid ash material was also produced and dropped in the hopper beneath the reactor tube. The ash consisted predominately of fused glass beads of various colors. This material was analyzed for residual carbonaceous material by pulverizing it and carrying out the same heating versus weight loss analysis performed on the original oil shale. No weight loss was observed upon heating from 932° to 1436°F., indicating that none of the organic material present in the original shale was left in the ash material. A 14% weight loss was observed upon heating the solid ash from 1436°F. to 2192°F., which indicated that most of the calcium carbonate present in the original sample remained in the ash and that some of this calcium carbonate had lost carbon dioxide during the reaction. Treating the ash with 0.1 N HCl resulted in the evolution of hydrogen sulfide and carbon dioxide, which indicated that whatever sulfur had been present in the original sample was at least in part found also in the ash.

The gaseous component of the effluent from the reactor was dried and then analyzed with a mass spectrometer. The results, reported in mole percent, were as follows: hydrogen (87.86%); methane (0.74%); acetylene (0.07%); ethylene (0.39%); nitrogen (1.24%); carbon monoxide (8.70%); mixed hydrocarbons (0.04%); carbon dioxide (0.016%); benzene (0.016%); toluene (0.002%); and

hydrogen sulfide (<0.0005%). This gas is suitable for use as a low-sulfur fuel.

EXAMPLE VI Steam Reforming And Gasification Of Sewage Sludge

A sample of activated sewage sludge, consisting of dried human waste admixed with siliceous clay binder and prilled to a particle size of approximately 2mm, was analyzed and found to have the following composition (Concentrations being expressed in weight percent): organic carbon (33.21%); organic hydrogen (4.38%); organic nitrogen (6.04%); organic sulfur (0.23%); water (6.14%); and inorganic residue (50%).

The sludge was introduced into the reactor at a rate of 54 pounds per hour. A total of 25 pounds was added. Steam at 250°F. was simultaneously fed into the reactor at 55 pounds per hour, which was about twice the stoichiometric rate for the water-gas reaction. Hydrogen was injected through the porous wall at a rate of 5 scfm. The temperature of the reactor was maintained at 3750°F.

The products of the reaction were a dense, white fog and a solid residue. The residue, which collected in a trap below the reactor tube, weighed 15 pounds and corresponded to 60% by weight of the activated sludge. The residue had the following composition (concentrations being expressed in weight percent): organic carbon (12.88%); organic hydrogen (1.69%); organic nitrogen (2.34%); organic sulfur (0.36%); water (trace); and inorganic residue (83%).

A portion of the vapor effluent from the reactor was condensed in a liquid nitrogen trap. The sample collected in the trap was brought to room temperature and found to have liquid and gaseous components. The boiling point of the liquid was 212°F., indicating that it was water. The gaseous component, which was suitable for use as a low-sulfur fuel, was analyzed with a mass spectrometer and gas chromatograph and found to have the following composition (concentrations being expressed as mole percent): hydrogen (60.933%); ammonia (0.0005%); methane (1.320%); water (0.083%); acetylene (0.463%); ethylene (0.304%); ethane (0.102%); hydrogen

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5 cyanide (0.281%); nitrogen (0.990%); carbon monoxide (34.122%); oxygen (0.0005%); argon (0.0078%); butene (0.175%); butane (0.026%); carbon dioxide (0.996%); benzene (0.100%); toluene (0.019%); hydrogen sulfide (0.0005%); and dicyanide (0.008%).

EXAMPLE VII

Partial Pyrolysis of Gas Oil

10 To demonstrate the use of the fluidwall reactor in the partial pyrolysis of petroleum distillates, a light lube stock or "gas oil" was partially pyrolyzed. This particular petroleum distillate was characterized by 15 the following distillation analysis:

Temperature (°F.)	% Distilled
174°	0
392°	10
428°	20
20 446°	30
482°	40
518°	50
532°	60
25 532°	70
536°	80
536°	90

30 The gas oil was introduced into the reactor tube in the form of a fog by atomizing it through a fogging nozzle. Hydrogen was employed as the atomizing gas as well as to form the fluid-wall. In addition, hydrogen was introduced into the inlet end of the reactor tube through a sweep gas inlet to sweep the gas oil fog through the tube.

35 The reactor tube was initially heated to 3400°F., with about 5 scfm of hydrogen being introduced into the plenum to form the fluid-wall and about 5 scfm of hydrogen being introduced into the sweep gas inlet.

40 The gas oil was then introduced into the reactor tube at about 0.25 gallon per minute, using about 5 scfm hydrogen for the atomizing gas. The temperature of the effluent stream just below the outlet of the reactor was set to about 820°F. by lowering the temperature of the reactor tube to 2600°F. Before samples were taken, the reactor was given time to stabilize at these operating conditions.

50 Samples of the effluent stream were collected by three methods, namely (1) by passing a portion of the effluent stream through a liquid nitrogen trap and collecting a sample by freezing it; (2) by 55 collecting gaseous samples from the stream at a position downstream from the liquid nitrogen trap; and (3) by passing a portion of the stream through a water-cooled condenser and collecting a liquid fraction. 60 The material collected in the liquid nitrogen trap was allowed to warm to about 50°F. and samples of the liquid and vapor

phases of this material at this temperature were then collected.

The liquid collected below the water-cooled condenser was characterized by the following distillation analysis:

Temperature (°F.)	% Distilled
257°	0
491°	10
543°	19
590°	29
617°	38
619°	48
648°	58
666°	67
691°	77
702°	87
734°	95

80 The liquid-phase sample collected from the liquid nitrogen trap was dried to remove water and was then analyzed and found to contain xylene, styrene, toluene, benzene, pentane, pentadiene, cyclopentadiene, butene, butadiene, propylene, methyl acetylene, methyl naphthalene, naphthalene, and higher molecular weight hydrocarbons. The gaseous component of the material collected in the liquid nitrogen trap was dried and analyzed with a mass spectrometer and gas chromatograph. After correcting for the presence of air, two samples of this gaseous component were found to have the following average composition (concentrations being expressed as mole percent): hydrogen (88.23%); methane (4.62%); ethylene (3.09%); propylene (1.22%); acetylene (0.55%); ethane (0.41%); butene (0.36%); benzene (0.35%); butadiene (0.31%); carbon dioxide (0.14%); pentadiene (0.13%); pentane (0.13%); propane (0.12%); carbon monoxide (0.12%); cyclopentadiene (0.10%); methyl pentadiene (0.06%); cyclohexane (0.03%); butane (0.03%); methyl acetylene (0.02%); and toluene (0.02%).

EXAMPLE VIII

Partial Steam Reforming Of Gas Oil

110 Gas oil identical to that used in Example VII was partially reformed with steam in the fluid-wall reactor in two substantially identical runs. In each of these runs, the gas oil was introduced into the reactor in the form of a fog by atomizing it through a fogging nozzle. Hydrogen was used for the fluid blanket, sweep gas, and atomizing gas at a rate of about 5 scfm for each purpose.

115 In both runs, the reactor tube was initially heated to 3300°F., with hydrogen being introduced into the sweep gas inlet and the plenum at approximately the rates to be used in the run. The gas oil was then introduced into the reactor at 120

approximately 0.25 gallon per minute together with steam at about 4 pounds per minute which corresponded to a carbon-to-steam molar ratio of about 1.0:1.6. Under the thermal load of the gas oil and steam, the temperature of the reactor fell to 2900°F. The temperature of the effluent stream just below the outlet was about 850°F. Samples were collected and treated in the same manner as in Example VII.

The liquid collected below the water-cooled condenser in the first run was characterized by the following distillation analysis:

	Temperature (°F.)	% Distilled
	482°	0
	581°	10
	617°	20
	635°	30
20	635°	40
	651°	50
	673°	60
	684°	70
	684°	80
25	716°	90

In the second run, a sample of the liquid component collected from the liquid nitrogen trap was warmed to 50°F., then dried to remove water, and then analyzed qualitatively. The resultant sample was found to contain toluene, benzene, pentene, pentadiene, cyclopentadiene, butene, butadiene, naphthalene, xylene, styrene, and higher molecular weight hydrocarbons. That portion of the original sample from the liquid nitrogen trap which was volatile at 50°F. was dried and analyzed with a gas chromatograph and mass spectrometer and was found to have the following composition after correcting for the presence of air (concentrations being expressed in mole percent): ethylene (36.85%); propylene (23.22%); acetylene (8.56%); ethane (7.99%); hydrogen (4.41%); butene (4.41%); butadiene (3.50%); propane (2.47%); methane (2.10%); methyl acetylene (1.98%); benzene (1.56%); pentadiene (0.62%); pentene (0.62%); cyclopentadiene (0.49%); carbon dioxide (0.37%); butane (0.25%); methyl pentadiene (0.25%); cyclohexan (0.13%); and toluene (0.04%).

EXAMPLE IX

Thermal Dissociation of Sawdust

Sawdust, a typical lignin-containing by-product, was thermally dissociated in the reactor tube 61 at a temperature of 3400°F. while hydrogen was forced through the porous wall of the tube at a rate of 5 scfm. The sawdust was fed into the reactor at a rate of about 50 pounds per hour. The pyrolysis products consisted of finely

divided carbon black similar to that produced by the dissociation of methane, gaseous products from the dissociation of volatile compounds, and an open-weave char in which the fibrous structure of the original wood was essentially intact.

EXAMPLE X

Silicon Carbide Abrasives From Silica

Silica sand, having a particle size distribution in the range from -50 to +100 mesh, was introduced into the reactor tube 61 through inlet 121 at a rate of 10 pounds per hour. Methane was simultaneously added to the reactor tube through inlet 91 at a rate of 1 scfm. The temperature of the reactor tube was held at 3400°F. Nitrogen was injected into the reactor tube through the porous wall at a rate of 5 scfm to form the fluid-wall. A powdered material dropped from the reactor tube and was collected in a hopper below.

The powdered product was sufficiently abrasive to scratch glass easily, indicating that it contained silicon carbide. Microscopic examination of the powder showed that it consisted of spheres of silicon dioxide covered with a shell composed of amorphous carbon and thin platelets of crystalline silicon carbide.

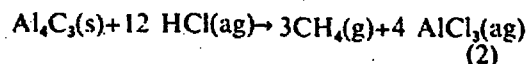
EXAMPLE XI

Production Of Aluminium Carbide

A stoichiometric mixture of aluminium powder and elemental carbon was prepared for the anticipated reaction:

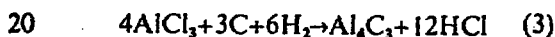


This mixture was introduced into the reactor at a rate of approximately 10 pounds per hour. Reactor tube 61 was maintained at 3400°F., and hydrogen was forced through the porous wall of the reactor tube at a rate of 5 scfm. The reaction yielded an amorphous, gray-brown material, which was collected in a trap below the reactor tube. A sample of the gray-brown product was mixed with 0.1 N HCl. A gas evolved which burned with the characteristic yellow flame of methane, which indicated that the following reaction had occurred between the product and the hydrochloric acid:



The sample dissolved completely in the hydrochloric acid, yielding a clear solution. Since the elemental carbon used as a starting material is insoluble in 0.1 N HCl, this indicated that the aluminium and carbon reacted quantitatively in the fluid-wall reactor to form aluminium carbide.

To test the feasibility of producing aluminium carbide in the fluid-wall reactor from aluminum chloride and carbon, anhydrous AlCl_3 was placed in a carbon crucible and heated until it sublimed. The aluminum chloride vapor was mixed into a stream of hydrogen and the resultant stream was then passed over a bed of carbon black. An arc-image lamp was focused on the surface of the carbon bed and heated an area of the bed to 1830°F ., as measured by an optical pyrometer. Small orange crystals formed just downstream from the heated zone, indicating that the aluminium chloride had reacted with carbon and hydrogen to produce aluminum carbide and hydrogen chloride in accordance with the following reaction:



When the orange crystals were added to 0.1 N HCl, the crystals dissolved in a gas was evolved which burned with the characteristic yellow flame of methane.

Since this procedure simulated what could be accomplished in the fluid-wall reactor of the invention by reacting aluminium chloride with carbon and hydrogen (produced by thermal dissociation of a gas or liquid hydrocarbon), it is evident that methane can be manufactured by (1) reacting aluminum chloride with an inexpensive hydrocarbonaceous material to form aluminum carbide and hydrogen chloride, and (2) quenching the reaction product in water so that the resultant aqueous hydrochloric acid hydrolyzes the aluminium carbide to produce methane and aluminium chloride which, in turn, can be recycled through the process.

EXAMPLE XII Reduction Of Ferric Oxide With Hydrogen

To demonstrate the utility of the fluid-wall reactor for reducing metal ores, pure ferric oxide (-100 mesh) was fed into the reactor at a rate of 35.1 pounds per hour at the same time as hydrogen was forced through the porous wall at a rate of 5 scfm. The hydrogen thus served both to form the fluid-wall and as the reducing agent for the iron oxide. The reactor tube was maintained at a temperature of 3400°F ., as measured by focusing an optical pyrometer on the incandescent inner wall of the tube. The temperature of the reactants in the reactor tube was determined to be 2750°F ., as measured with the optical pyrometer. A gray powder was produced which collected in the hopper beneath the reactor tube. The

temperature of the effluent stream just below the outlet of the reactor was measured at 600°F .

The product was pure iron powder, which tended to be pyrophoric at temperatures of about 300°F ., when freshly prepared. Viewing the powder with a microscope showed that it consisted of small, spherical particles, which indicated that the iron had been in a molten state during its passage through the reactor tube.

EXAMPLE XIII Thermal Dissociation Of Hydrogen Sulfide And Methane

Using the fluid-wall reactor, hydrogen sulfide was reacted with the *in situ* carbon formed by the thermal dissociation of methane, thereby forming carbon disulfide and hydrogen. Runs were performed at two different temperatures, namely at 2975°F ., and at 3200°F .. In both instances, temperatures were measured by focusing an optical pyrometer on the incandescent reactants in the reactor tube, the carbon particles from the dissociating methane being the primary incandescent constituents of the reaction mixture. Hydrogen was forced through the porous wall of the reactor tube at a rate of 5 scfm to serve as the blanket gas. Hydrogen sulfide at a rate of 0.32 scfm and methane at a rate of 1 scfm were mixed together and introduced into the reactor tube. The gas mixture was at room temperature at the inlet to the reactor tube. A target of carbon black was added to initiate the reaction, although once the reaction was initiated it was self-sustaining and no further carbon black was needed.

Samples of the gaseous component of the products for the two runs were analyzed with a mass spectrometer. The results of the analysis are given in the following table, the concentrations being reported in mole percent:

Compound	Reaction Temperature	
	2975°F .	3200°F .
Hydrogen	83.974	88.560
Methane	11.379	6.230;
Acetylene	1.681	2.281
Ethylene	1.397	1.519
Hydrogen sulfide	1.021	0.813
Carbon dioxide	0.296	0.160
Carbon disulfide	0.216	0.403
Benzene	0.036	0.034

Although each of the foregoing examples was conducted in the fluid-wall reactor shown in Figures 1A and 1B (the first embodiment), even better results can be achieved by using the fluid-wall reactor of Figures 6A to 6D with suitable modifications (where necessary) to handle

solid feedstocks. The use of process variable control systems should permit the optimum operating conditions to be located and maintained accurately. If such control systems incorporate a digital computer, the search for the optimum operating conditions can be carried out automatically.

WHAT WE CLAIM IS:—

1. A process for carrying out a chemical reaction at an elevated temperature, wherein radiation is caused to be incident on one or more reactants situated in a reaction zone defined by a wall of fluid substantially transparent to that radiation, the wall of fluid being located within a heat shield of refractory material which reflects radiation, sufficient radiant energy being absorbed in the reaction zone to raise the temperature of the or at least one reactant to sustain the chemical reaction.
2. A process as claimed in Claim 1, wherein radiation is absorbed by the or at least one reactant.
3. A process as claimed in Claim 1 or Claim 2, wherein radiation is absorbed by a target within the reaction zone and energy is transferred from the target to the or at least one reactant.
4. A process as claimed in any one of Claims 1 to 3, wherein the fluid forming the wall of the reaction zone is inert with respect to the reaction being carried out.
5. A process as claimed in any one of Claims 1 to 4, wherein the or each reactant is passed continuously through the reaction zone from an inlet to an outlet.
6. A process as claimed in any one of Claims 1 to 5, wherein the fluid wall defining the reaction zone is in the form of an envelope of generally annular cross-section situated within a reactor tube having an inlet end and an outlet end, the reactor tube being positioned within the heat shield.
7. A process as claimed in Claim 6, wherein the reactor tube is made of porous material and the envelope of fluid is generated by causing the fluid to flow radially inwardly through the porous tube wall.
8. A process as claimed in Claim 7, wherein the reactor tube is enclosed within a fluid-tight pressure vessel containing a plenum of fluid the inner boundary of which is defined by the outer wall of the reactor tube the inlet and outlet ends of the tube being sealed from the plenum and the fluid being admitted to the plenum through an inlet and being introduced under pressure into the reactor tube from the plenum through the porous tube wall.
9. A process as claimed in any one of Claims 6 to 8, wherein the reactor tube is

made of refractory material and its temperature is so raised that it emits the radiation that serves to raise the temperature of the or at least one reactant to sustain the chemical reaction.

10. A process as claimed in Claim 9, wherein the reactor tube is heated by electrical heating means situated outside the tube.

11. A process as claimed in Claim 10, wherein the heat shield is circumferential and is so situated within the pressure vessel outside the electrical heating means as to reflect radiation from the electrical heating means towards the tube.

12. A process as claimed in any one of Claims 7 to 11, wherein the reactor tube is made of fabric or a fibrous refractory material.

13. A process as claimed in Claim 12, wherein prior to the process the fabric is stiffened by the deposition of a refractory coating onto it.

14. A process as claimed in Claim 13, wherein the refractory coating comprises pyrolytic graphite.

15. A process as claimed in any one of Claims 12 to 14, wherein prior to or during the process the pores of the fabric are enlarged by means of an etching agent carried by the fluid defining the reaction zone.

16. A process as claimed in claim 15, wherein the etching agent comprises steam or molecular oxygen.

17. A process as claimed in any one of Claims 12 to 16, wherein prior to or during the process the pores of the fabric are reduced in diameter by means of a refractory deposition agent carried by the fluid defining the reaction zone.

18. A process as claimed in Claim 17, wherein the refractory deposition agent is carbonaceous gas.

19. A process as claimed in any one of Claims 12 to 18, wherein prior to the process the fibrous refractory material of the tube wall is at least partially coated with a refractory oxide.

20. A process as claimed in Claim 19, wherein the coating is effected by treating the reactor tube with a volatile metal-containing compound.

21. A process as claimed in any one of Claims 5 to 20, wherein the reaction product(s) and any remaining reactant(s) are cooled immediately after they have left the reaction zone.

22. A process as claimed in Claim 21, wherein cooling is effected by radiation heat transfer to a cool radiant-energy absorbing surface.

23. A process as claimed in any one of Claims 1 to 22, wherein the or each reactant is additionally heated, at least initially, to

solid feedstocks. The use of process variable control systems should permit the optimum operating conditions to be located and maintained accurately. If such control systems incorporate a digital computer, the search for the optimum operating conditions can be carried out automatically.

WHAT WE CLAIM IS:—

1. A process for carrying out a chemical reaction at an elevated temperature, wherein radiation is caused to be incident on one or more reactants situated in a reaction zone defined by a wall of fluid substantially transparent to that radiation, the wall of fluid being located within a heat shield of refractory material which reflects radiation, sufficient radiant energy being absorbed in the reaction zone to raise the temperature of the or at least one reactant to sustain the chemical reaction.

2. A process as claimed in Claim 1, wherein radiation is absorbed by the or at least one reactant.

3. A process as claimed in Claim 1 or Claim 2, wherein radiation is absorbed by a target within the reaction zone and energy is transferred from the target to the or at least one reactant.

4. A process as claimed in any one of Claims 1 to 3, wherein the fluid forming the wall of the reaction zone is inert with respect to the reaction being carried out.

5. A process as claimed in any one of Claims 1 to 4, wherein the or each reactant is passed continuously through the reaction zone from an inlet to an outlet.

6. A process as claimed in any one of Claims 1 to 5, wherein the fluid wall defining the reaction zone is in the form of an envelope of generally annular cross-section situated within a reactor tube having an inlet end and an outlet end, the reactor tube being positioned within the heat shield.

7. A process as claimed in Claim 6, wherein the reactor tube is made of porous material and the envelope of fluid is generated by causing the fluid to flow radially inwardly through the porous tube wall.

8. A process as claimed in Claim 7, wherein the reactor tube is enclosed within a fluid-tight pressure vessel containing a plenum of fluid the inner boundary of which is defined by the outer wall of the reactor tube the inlet and outlet ends of the tube being sealed from the plenum and the fluid being admitted to the plenum through an inlet and being introduced under pressure into the reactor tube from the plenum through the porous tube wall.

9. A process as claimed in any one of Claims 6 to 8, wherein the reactor tube is

made of refractory material and its temperature is so raised that it emits the radiation that serves to raise the temperature of the or at least one reactant to sustain the chemical reaction.

10. A process as claimed in Claim 9, wherein the reactor tube is heated by electrical heating means situated outside the tube.

11. A process as claimed in Claim 10, wherein the heat shield is circumferential and is so situated within the pressure vessel outside the electrical heating means as to reflect radiation from the electrical heating means towards the tube.

12. A process as claimed in any one of Claims 7 to 11, wherein the reactor tube is made of fabric or a fibrous refractory material.

13. A process as claimed in Claim 12, wherein prior to the process the fabric is stiffened by the deposition of a refractory coating onto it.

14. A process as claimed in Claim 13, wherein the refractory coating comprises pyrolytic graphite.

15. A process as claimed in any one of Claims 12 to 14, wherein prior to or during the process the pores of the fabric are enlarged by means of an etching agent carried by the fluid defining the reaction zone.

16. A process as claimed in claim 15, wherein the etching agent comprises steam or molecular oxygen.

17. A process as claimed in any one of Claims 12 to 16, wherein prior to or during the process the pores of the fabric are reduced in diameter by means of a refractory deposition agent carried by the fluid defining the reaction zone.

18. A process as claimed in Claim 17, wherein the refractory deposition agent is carbonaceous gas.

19. A process as claimed in any one of Claims 12 to 18, wherein prior to the process the fibrous refractory material of the tube wall is at least partially coated with a refractory oxide.

20. A process as claimed in Claim 19, wherein the coating is effected by treating the reactor tube with a volatile metal-containing compound.

21. A process as claimed in any one of Claims 5 to 20, wherein the reaction product(s) and any remaining reactant(s) are cooled immediately after they have left the reaction zone.

22. A process as claimed in Claim 21, wherein cooling is effected by radiation heat transfer to a cool radiant-energy absorbing surface.

23. A process as claimed in any one of Claims 1 to 22, wherein the or each reactant is additionally heated, at least initially, to



raise its temperature sufficiently to initiate the reaction.

24. A process as claimed in Claim 23, wherein the or each reactant is additionally heated by means of an electrical heating element.

25. A process as claimed in Claim 23, wherein the or each reactant is additionally heated by means of an electric arc.

26. A process as claimed in Claim 23, wherein the or each reactant is additionally heated by means of a flame.

27. A process as claimed in Claim 3 or any claim appendant thereto, wherein the target comprises a target material which is passed through the reaction zone.

28. A process as claimed in Claim 27, wherein the target material is introduced into the reaction zone prior to the introduction of the reactants.

29. A process as claimed in Claim 27 or Claim 28, wherein the target material is cooled immediately after leaving the reaction zone.

30. A process as claimed in any one of Claims 27 to 29, wherein the introduction of the target material is discontinued after the reaction has been initiated.

31. A process as claimed in any one of Claims 27 to 30, wherein the target material is a liquid.

32. A process as claimed in any one of Claims 27 to 30, wherein the target material is gas that absorbs electromagnetic radiation having a wavelength within the range of from 0.01 to 100 μm .

33. A process as claimed in any one of Claims 27 to 30, wherein the target material is finely divided carbon powder.

34. A process as claimed in Claim 3 or any claim appendant thereto, wherein the reaction zone contains a solid target member.

35. A process as claimed in Claim 34, wherein the target member is made of carbon.

36. A process as claimed in any one of Claims 1 to 35, wherein the power flux in the reaction zone is at least 180 watts/cm².

37. A process as claimed in Claim 6 or any claim appendant thereto, wherein the temperature of the reactor tube is at least 2300°F.

38. A process as claimed in any one of Claims 1 to 37, wherein at least one hydrocarbon and/or at least one hydrocarbonaceous material is dissociated into hydrogen and carbon black.

39. A process as claimed in any one of Claims 1 to 37, wherein at least one carbonaceous or hydrocarbonaceous material is steam-reformed to yield a gaseous product comprising carbon monoxide and hydrogen.

40. A process as claimed in Claim 39,

wherein the carbonaceous or hydrocarbonaceous material is selected from coal, petroleum fractions, oil shale, tar sands and lignite.

41. A process as claimed in Claim 39 or Claim 40, wherein the starting material additionally comprises one or more inorganic carbonates and/or inorganic oxides in order to facilitate the removal of sulphur-containing contaminants from the said gaseous product.

42. A process as claimed in any one of Claims 1 to 37, wherein at least one hydrocarbon and/or at least one hydrocarbonaceous material is partially dissociated into lower-molecular-weight compounds.

43. A process as claimed in any one of Claims 1 to 37, wherein at least one saturated hydrocarbon is subjected to partial pyrolysis to yield one or more unsaturated hydrocarbons.

44. A process as claimed in any one of Claims 1 to 37, wherein organic waste material is converted into a fuel gas.

45. A process as claimed in any one of Claims 1 to 37, wherein a catalyst for promoting the formation of free radicals, carbonium ions or carbanions is added to the reactant or reactants.

46. A process as claimed in any one of Claims 1 to 37, in which a sulphur containing carbonaceous or hydrocarbonaceous material is subjected to partial or complete desulphurisation.

47. A process as claimed in any one of Claims 1 to 37, wherein a mineral ore and/or an inorganic compound is reduced by means of a reducing agent to the element or to a compound in which the element is in a lower oxidation state.

48. A process as claimed in Claim 47, wherein the reducing agent is hydrogen, carbon, carbon monoxide, or a gas mixture comprising carbon monoxide and hydrogen.

49. A process as claimed in any one of Claims 1 to 37, wherein an inorganic element or compound is partially or completely reacted with a carbonaceous or hydrocarbonaceous material to yield an inorganic carbide.

50. A process as claimed in Claim 49, wherein aluminium chloride is reacted with a hydrocarbon or a hydrocarbonaceous material to yield a product comprising aluminium carbide and hydrogen chloride, and the said product is quenched with water to yield methane and aluminium chloride.

51. A process as claimed in Claim 50, wherein the aluminium chloride thus formed is recycled into the process as a reactant.

52. A process for carrying out a chemical



reaction at an elevated temperature, carried out substantially as hereinbefore described with reference to Figures 1A to 5; Figures 6A to 11, Figure 12, Figures 13A and 13B, Figure 14, Figure 15, Figure 16 or Figure 18 of the accompanying drawings.

53. A process for carrying out a chemical reaction at an elevated temperature, carried out substantially as described in any one of the Examples herein.

54. A product of a high-temperature chemical reaction whenever produced by a process as claimed in any one of Claims 1 to 53.

55. A reactor for carrying out a chemical reaction at an elevated temperature, comprising a reactor tube having an inlet end and an outlet end, means for generating an envelope of fluid to define a fluid-walled reaction zone within the reactor tube, and means for causing, in operation, sufficient radiant energy to be incident upon the reaction zone to raise the temperature of one or more reactants situated there to sustain a chemical reaction, the reactor tube being located within a heat shield of refractory material capable of reflecting the said radiant energy.

56. A reactor as claimed in Claim 55, which includes means situated outside the reactor tube adjacent to its outlet for cooling a product and/or a reactant leaving the reactor tube.

57. A reactor as claimed in Claim 55 or Claim 56, which includes additional heating means within the reactor tube for raising the temperature of the reactant or reactants to initiate the reaction.

58. A reactor as claimed in Claim 57, wherein the additional heating means comprises an electrical heating element, an electric arc generator, or a flame burner.

59. A reactor as claimed in any one of Claims 55 to 59, wherein a radiant-energy-absorbing target member is positioned within the reaction zone.

60. A reactor as claimed in Claim 59, wherein the target member is made of carbon.

61. A reactor as claimed in any one of Claims 55 to 60, wherein the reactor tube is made of porous material, and the means for generating an envelope of fluid within the reactor tube comprises means for introducing a fluid under pressure through the porous tube wall.

62. A reactor as claimed in Claim 67, wherein the pore diameter of the reactor tube is within the range of from 0.001 to 0.020 inch.

63. A reactor as claimed in Claim 58, wherein the reactor tube is enclosed within a fluid-tight pressure vessel containing a region for fluid the inner boundary of which is defined by the outer wall of the

reactor tube, the inlet and outlet ends of the tube being sealed from the region and the region having an inlet for fluid, the pores of the tube being of such a diameter as to permit a substantially uniform flow of sufficient fluid from the region through the tube wall.

64. A reactor as claimed in claim 63, which further includes means for cooling the pressure vessel.

65. A reactor as claimed in Claim 64, wherein the cooling means comprises cooling coils disposed about the outer wall of the pressure vessel.

66. A reactor as claimed in any one of Claims 59 to 64, wherein the porous material of the tube is refractory and the reactor further comprises electrical heating means positioned within the said region to raise the temperature of the reactor tube.

67. A reactor as claimed in Claim 66, wherein the heat shield is positioned within the pressure vessel radially outwardly of the heating means, for reflecting radiant energy from the heating means towards the reaction zone.

68. A reactor as claimed in Claim 67, wherein the heat shield is made of molybdenum or a graphite material.

69. A reactor as claimed in any one of Claims 62 to 64, wherein the electrical heating means comprises a plurality of electrically resistive heating elements disposed radially outwardly of, and spaced circumferentially about, the reactor tube.

70. A reactor as claimed in Claim 69, wherein the resistive heating elements are made of a fabric or a fibrous refractory material.

71. A reactor as claimed in Claim 70, wherein the fibrous refractory material comprises graphite or another form of carbon.

72. A reactor as claimed in any one of Claims 66 to 71, wherein the reactor tube is made of graphite or another form of carbon, sintered stainless steel, sintered tungsten, sintered molybdenum, thorium oxide, magnesium oxide, zinc oxide, aluminium oxide or zirconium oxide.

73. A reactor as claimed in any one of Claims 66 to 71, wherein the reactor tube is made of a fabric or a fibrous refractory material.

74. A reactor as claimed in Claim 73, wherein the fibrous refractory material is graphite or another form of carbon.

75. A reactor as claimed in Claim 73 or Claim 74, which further includes means for depositing a refractory coating upon the fibrous refractory material of the reactor tube to increase the rigidity of the fabric.

76. A reactor as claimed in Claim 75, wherein the refractory coating depositing means includes means for opening the



region for containing fluid to exhaust, means for introducing a sweep gas into the reactor tube and thence into the said region, metering means for dispensing a refractory deposition agent into the sweep gas stream, and means for closing the reactor tube outlet, the arrangement being such that the sweep gas containing the deposition agent is, in operation, directed into the reaction zone and radially outwardly through the reactor tube wall into the region for containing fluid.

77. A reactor as claimed in any one of Claims 73 to 76, which further includes means for enlarging the diameter of the pores in the fabric of the tube wall in order to increase the flow of fluid through the tube wall.

78. A reactor as claimed in Claim 77, in which the means for enlarging the diameter of the pores includes one or more sensors to determine the pressure differential between the region for containing fluid and the interior of the reactor tube, and metering means for dispensing an etching agent into the fluid stream.

79. A reactor as claimed in any one of Claims 73 to 78, which further includes means for reducing the diameter of the pores in the fabric of the tube wall in order to decrease the flow of fluid through the tube wall.

80. A reactor as claimed in Claim 79, wherein the means for reducing the diameter of the pores includes one or more sensors to determine the pressure differential between the region for containing fluid and the interior of the reactor tube, and metering means for dispensing a refractory deposition agent into the fluid stream.

81. A reactor as claimed in any one of Claims 73 to 80, wherein the fibrous refractory material of the tube wall is at least partially coated with a refractory oxide.

82. A reactor as claimed in Claim 81, wherein the refractory oxide is thorium oxide, magnesium oxide, zinc oxide, aluminium oxide or zirconium oxide.

83. A reactor as claimed in Claim 63 or any claim appendant thereto, which further includes a tubular bellows disposed within an inlet assembly section of the pressure vessel, one end of the bellows being secured in a fluid-tight manner to the inlet assembly section and the other end of the bellows being secured in a fluid-tight manner to the inlet end of the reactor tube by a reactor tube inlet support ring, the bellows being deformable to accommodate axial expansion and contraction of the reactor tube.

84. A reactor as claimed in any one of Claims 55 to 83, which further includes

means for applying an axial tensile force to the reactor tube.

85. A reactor as claimed in any one of Claims 55 to 84, which includes means for introducing a liquid reactant into the reaction zone of the reactor tube, the means comprising a fogging nozzle disposed within the reactor tube adjacent to an inlet of the reaction zone, the fogging nozzle being arranged to mix the liquid reactant and an atomizing gas under pressure within the nozzle and to disperse the thus formed mixture from the nozzle outlet into the reaction zone as a fog.

86. A reactor as claimed in Claim 85, wherein the fogging nozzle includes a tubular shroud secured to and disposed radially outwardly of the nozzle, the axis of the shroud being substantially parallel to the axis of the reactor tube.

87. A reactor as claimed in Claim 85 or Claim 86, which includes a plurality of fogging nozzles disposed within the reactor tube adjacent to the inlet end of the reaction zone.

88. A reactor as claimed in any one of Claims 85 to 87, wherein the means for introducing a liquid reactant into the reaction zone further includes means for introducing a sweep gas into the inlet end of the reactor tube to direct a fog of liquid reactant from the fogging nozzle towards the reaction zone.

89. A reactor as claimed in any one of Claims 55 to 88, wherein a region of the interior of the reactor tube between the inlet end of the tube and the reaction zone defines a pre-reaction zone into which fluid may be directed to form a protective blanket to assist in confining a reactant or reactants within the reaction zone out of contact with the inner wall of the reactor tube.

90. A reactor as claimed in any one of Claims 55 to 89, which includes means for introducing a solid reactant into the reaction zone of the reactor tube, the means comprising a helical feed screw rotatably mounted within an elongated tubular housing, drive means for rotating the feed screw, a hopper for introducing a crushed, solid reactant into the housing, means for introducing a pressure sealing fluid into the housing at a point downstream from the hopper, an outlet means for discharging the reactant and the sealing fluid from the housing into the reactor inlet.

91. A reactor as claimed in any one of Claims 55 to 90, which further comprises a counterflow heat exchanger of non-uniform temperature profile and having an inlet end and an outlet end, the heat exchanger being securable at its inlet end to the outlet of the reactor tube in



- order to receive high temperature reaction products, the heat exchanger comprising an inner tubular wall of refractory material, an outer tubular wall of refractory material spaced coaxially outwardly from the inner wall, and a spiral baffle of refractory material disposed between the inner and outer walls to define a spiral, annular coolant channel, the channel having at least one inlet extending through the outer wall in communication with the channel and an outlet adjacent to the inlet end of the heat exchanger.
92. A reactor as claimed in Claim 91 as appendant to Claim 63, wherein the heat exchanger further includes means for directing coolant from the outlet of the spiral, annular channel into the region of the reactor for containing fluid.
93. A reactor as claimed in Claim 66 or any claim appendant thereto, which further comprises a reaction product control system comprising:
- (i) means for withdrawing samples of reaction product leaving the reactor;
 - (ii) a reaction product analyser including a sample inlet and a signal output, the analyzer being arranged to compare the chemical composition of the reaction product to a preselected composition and to generate an electrical signal at its output corresponding to deviations in the chemical composition of samples being analyzed; and
 - (iii) a reactor temperature controller including a control signal input connected to the analyzer signal output and a heater power output connected to the electrical heating means for the reactor tube, the temperature of the reactor tube being, in operation, so variable in response to changes in the analyser signal as to reduce the deviations.
94. A reactor as claimed in Claim 93, wherein the reaction product analyser comprises a gas chromatograph connected to a digital computer.
95. A reactor as claimed in Claim 93 or Claim 94, wherein the means for withdrawing samples comprises means for transferring samples to the sample inlet of the reaction product analyser at preselected time intervals.
96. A reactor as claimed in any one of Claims 93 to 95, wherein the reactor temperature controller includes an SCR circuit connected in series with a three phase power line.
97. A reactor for carrying out a chemical reaction at an elevated temperature, substantially as hereinbefore described with reference to, and as shown in, Figures 1A to 5 of the accompanying drawings.
98. A reactor for carrying out a chemical reaction at an elevated temperature, substantially as hereinbefore described with reference to, and as shown in Figures 6A to 11 of the accompanying drawings.
99. A reactor for carrying out a chemical reaction at an elevated temperature, substantially as hereinbefore described with reference to, and as shown in Figures 6A to 11 and 12 of the accompanying drawings.
100. A reactor for carrying out a chemical reaction at an elevated temperature, substantially as hereinbefore described with reference to, and as shown in Figures 6A to 11, 13A and 13B of the accompanying drawings.
101. A reactor for carrying out a chemical reaction at an elevated temperature, substantially as hereinbefore described with reference to, and as shown in Figures 6A to 11, 13A, 13B and 14 of the accompanying drawings.
102. A reactor for carrying out a chemical reaction at an elevated temperature, substantially as hereinbefore described with reference to, and as shown in Figures 6A to 11 and 15 of the accompanying drawings.
103. A reactor for carrying out a chemical reaction at an elevated temperature, substantially as hereinbefore described with reference to, and as shown in Figures 6A to 11 and 16 of the accompanying drawings.
104. A reactor for carrying out a chemical reaction at an elevated temperature, substantially as hereinbefore described with reference to, and as shown in Figures 6A to 11 and 18 of the accompanying drawings.
105. A high-temperature chemical reaction process whenever carried out using a reactor as claimed in any one of Claims 55 to 104.
106. A product of a high-temperature chemical reaction process as claimed in Claim 105.

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