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(71) Applicant *(for all designated States except US)*: **NATIONAL INSTITUTE OF CLEAN-AND-LOW-CARBON ENERGY** [CN/CN]; ShenHua Low Carbon 001# Post Box, Future Science & Techonology Park, Xiaotangshan, Changping District, Beijing 102209 (CN).

(72) Inventors; and

(75) Inventors/Applicants *(for US only)*: **CHENG, Yi** [CN/CN]; ShenHua Low Carbon 001# Post Box, Future Science & Techonology Park, Xiaotangshan, Changping District, Beijing 102209 (CN). **GUO, Yi** [US/CN]; ShenHua Low Carbon 001# Post Box, Future Science & Techonology Park, Changping District, Beijing 102209 (CN). **YAN, Binhang** [CN/CN]; ShenHua Low Carbon 001# Post Box, Future Science & Techonology Park, Xiaotangshan, Changping District, Beijing 102209 (CN). **WU, Changning** [CN/CN]; ShenHua Low Carbon 001# Post Box, Future Science & Techonology Park, Xiaotangshan, Changping District, Beijing 102209 (CN). **LI, Xuan** [CN/CN]; ShenHua Low Carbon 001# Post Box, Future Science & Techonology Park, Xiaotangshan, Changping District, Beijing 102209 (CN).

(74) Agent: **ZHONGZI LAW OFFICE**; 7F, New Era Building, 26 Pinganli Xidajie, Xicheng District, Beijing 100034 (CN).

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(54) Title: MULTI-STAGE PLASMA CRACKING CARBONACEOUS MATERIAL REACTOR AND PROCESS FOR PRODUCING ACETYLENE BY USING THE SAME

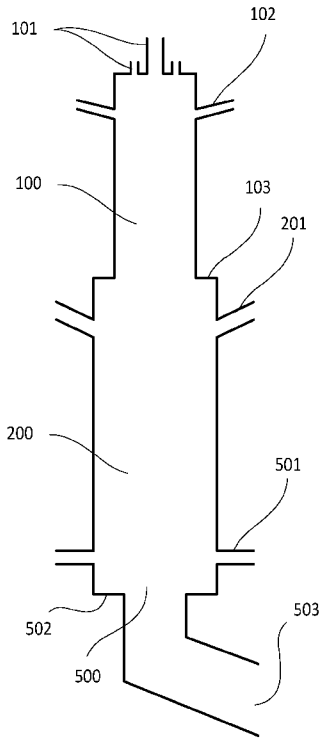


FIGURE 1

(57) Abstract: Disclosed is a multi-stage plasma reactor for cracking carbonaceous material comprising: first stage of reaction tube mainly for mixing of the carbonaceous material, carrier gas with first heating gas and pyrolysis of the carbonaceous material; second stage-N stage of reaction tube mainly for gas phase reaction of volatiles resulted from the pyrolysis, wherein N is integer of more than or equal to 2; at least one inlet of the carbonaceous material and carrier gas as feedstock located on top of the first stage of reaction tube; at least one inlet of first heating gas located on side surface of the first stage of reaction tube; at least one inlets of second-N heating gases located on side surface of the second stage-N stage of reaction tube respectively, wherein said second-N heating gases are plasma high temperature gases; at least one inlet of quench media for quenching or freezing of reaction products; at least one outlet of quenched products and gases located on bottom or lower portion of last stage of reaction tube.

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Multi - Stage Plasma Cracking Carbonaceous Material Reactor and Process for Producing Acetylene by Using the Same

FIELD OF INVENTION

The invention relates to an energy efficient equipment and process for cracking carbonaceous material with volatiles content to produce a high yield of cracked products, especially to a multi-stage plasma reactor for cracking carbonaceous material as well as a process for cracking carbonaceous material by using the multi-stage plasma reactor, more particularly, also to a process for producing acetylene by using the multi-stage plasma reactor.

PRIOR ART

Conventionally, carbonaceous materials, together with other components such as hydrogen, are heated by an electric arc device or other suitable sources of heat which are well known to those skilled in the art so as to be cracked or pyrolyzed. The composition that is produced as a result of the decomposition of the carbonaceous matter will depend on the reaction conditions existing in the decomposition or reaction zone. It is well-known that the formation of certain compositions is favored under specified reaction conditions. For example, the formation of acetylene as an intermediate product is favored where the temperature of the reaction zone is above 1300 K. The formation of acetylene relative to the simultaneous decomposition of acetylene is also favored at or near 1300 K.

Generally, where an electric arc is applied as a heat source, the electric arc passes through the gas, for example hydrogen gas, causing temperature of the gas to increase to extremely high temperatures in a very short time. Arc column temperatures commonly reach 8,000 K to 20,000 K. The gas on leaving the arc is commonly within the neighborhood of 2,000-5,000 K. Under these conditions, the gas molecule such as hydrogen molecules may dissociate partially into hydrogen atoms even H^+ or H^-, therefore generating plasma high temperature gas.

Once plasma high temperature gas, such as plasma hydrogen, leaves the electric arc, there is an extremely rapid tendency for the plasma gas atoms or ions, for example hydrogen atoms to recombine into molecules, and if doing so, they give off tremendous amounts of heat. A portion of this heat, in addition to the sensible heat of the plasma gas, is absorbed by the carbonaceous material particles which are adjacent to or contact with the plasma gas atoms or ions, mostly via conduction, convection and radiation, thereby causing the carbonaceous material particle to be pyrolyzed and/or cracked or compose and more specifically to give off its volatile content, i.e., to devolatilize.

It is also well-known and established that steps and conditions of which the above decomposition and devolatilization of the carbonaceous material will vary greatly with the type of the carbonaceous materials. Heretofore, gaseous and liquid carbonaceous materials were the favored feedstocks, as there was no known way of producing a high yield of cracked products, for example acetylene, at reasonable costs from solid carbonaceous materials. Also gaseous and liquid feedstocks were easier to be processed, and produced less wear and tear on the arc apparatus.

On the other hand though the basic process steps are known, it is possible that the mechanics and the kinetics of the process, for example, were not understood well enough, heretofore, to teach one how to maximize the yield of some specific cracked products, for example acetylene from solid carbonaceous matter in an energy efficient manner.

There have been a lot of attempts and experiments on the improvements of devices and processes aimed to maximize the yield of some specific cracked products from solid carbonaceous matter in the prior art.

For example, US4358629 disclosed a method of conversion by way of decomposing a solid carbonaceous matter to acetylene. Specifically, this patent taught selecting the operating conditions which will produce high yield at low cost. In this patent, specific

values of heat and enthalpy for the carbonaceous matter and the gas are proposed in combination with specific particle sizes and reaction time. All of the foregoing contributes to producing acetylene at a commercially competitive cost.

In fact, US4358629 described an electric arc reactor including four zones in turn along with the solid carbonaceous material motion direction, i.e., the solid carbonaceous material powder dispersion zone, arc zone, reaction zone, and quench zone. Because of the ultra-short residence time of the powder in the arc zone and the temporary thermal inertia of the powder at that time, temperature of the powder kept certainly close to its inlet temperature while the gas going through reached a high temperature up to 8000 K. The solid carbonaceous material powder could only be heated in the reaction zone by the heated stream through conduction and convection. In this way, all electricity input from the thin arc zone, i.e. a large amount of energy enough to raise the powder temperature to above 1800 K, led to unreasonable over-aggregation of energy and inevitable exposure of over-concentrated heat to the reactor wall thereby causing overheating of the wall. The heat essentially removed from the neighbor of the wall for protection of the wall accounts for about half of the total electricity input, as a result, a lot of valuable energy had to waste out. Furthermore, the extreme high temperature occurrence on the specific region greatly challenged the design of the reactor wall structure, selection of the wall material, as well as made the wall protection in big troubles.

Pyrolysis of Coal in Hydrogen and Helium Plasma (Baumann, H., Bittner, D., Beiers, H.G., Klein, J. & Juntgen, H, Fuel, 1988, Vol. 67, pp 1120-1123, August) and Pyrolysis of Some Gaseous and Liquid Hydrocarbons in Hydrogen Plasma (Beiers, H.G., Baumann, H., Bittner, D., Klein, J. and Juntgen, H, Fuel, 1988, Vol. 67, pp 1012-1016, July) disclosed one apparatus, consisting of a plasma generator and a plasma reactor, which was described to carry out the pyrolysis of coal or gaseous and liquid hydrocarbons. In this apparatus, high-temperature stream is generated in a plasma generator with a mean temperature of 3300 K at outlet, and then fed into the

reaction tube as reactor from its top entrance. The dried coal powder or gaseous and liquid hydrocarbons are injected into the reaction tube from its side entrance close to the top entrance, where the cold coal powders was estimated to be well mixed with the above hot plasma jet. However, due to the high-speed moving downward of the plasma jet and thereby forming strong obstacle to fluid-powder mixing, so that the contact and thermal transfer efficiency between the coal powder and plasma stream is weakened with negative effect on the reactor performances while such reactor structure and arrangement of feeding caused the caking phenomenon hardly be avoided due to the coal powder or gaseous and liquid hydrocarbons continuously flushing and striking on the wall surface.

CN1562922 disclosed a reactor similar to that described in the above articles but introduced argon gas sprayed onto inner wall of the reaction tube so as to prevent the inner wall of reaction tube from caking. However, the reactor disclosed in this patent document has still not overcome all of the above identified shortcomings.

US4536603 disclosed a process wherein coal was reacted with a hot gas stream to produce acetylene. The process comprised the sequential steps of reacting a fuel, oxygen and steam under controlled conditions of temperature to produce a hot gas stream principally comprising hydrogen, carbon monoxide and steam along with minor amounts of carbon dioxide, and essentially free of O, OH and O_2. The hot gas stream is accelerated to a high velocity and impinged upon a stream of particulate bituminous or sub-bituminous coal and thereafter the mixture of hot gas and coal is decelerated to a velocity of from about 150 to 300 feet/second. The amounts of the streams of particulate coal and hot gas are controlled to produce in the reaction zone a pressure in the range of from about 10 to 100 Pisa and a temperature of from about 1800 to 3000° F. The mixture of coal and hot gas is maintained at that pressure and temperature for a time of from about 2 to 30 milliseconds to produce a product stream including char and acetylene. The temperature of the product stream is then reduced to less than about 900° F. in a time of less than about 2 milliseconds to substantially

arrest any further reactions and the acetylene is recovered therefrom. The char is recovered and used as at least a part of the fuel used to produce hot gas.

US 4588850 disclosed a method for manufacturing acetylene and synthesis or reduction gas from coal by means of an electric arc or plasma process, wherein coal converted into powder form is pyrolyzed in an electric arc reactor with an energy density of 1 to 5 kWh/Nm^3, a residence period of 0.5 to 10 millisecond and at a temperature of at least 1500℃ such that amount of the gaseous compounds derived from the coal do not exceed 1.8 times of that of the so-called volatile content of the coal. The coke remaining after subsequent quenching is then fed to a second electric arc reactor in which the coke, by means of a gasifying medium in conjunction with heating by means of an electric arc or plasma process, is converted into synthesis or reduction gas with a residence period of 1 to 15 sec and at a temperature of at least 800℃. The gas flow from the pyrolysis zone is cleaned and acetylene is recovered therefrom by selective solvents. The gas from the cleaning step is similarly cooled and cleaned

CN101742808 disclosed a high-power V-shaped plasma generator being capable of displacing the conventional line-shaped plasma generator, and proclaimed to have relatively low energy consumption and convenient operation conditions. The V-shaped plasma generator could be applied to generate variety of plasma high temperature gases, for example plasma hydrogen and inert gases.

US4367363 disclosed a process related to the recovering of pure acetylene from the gaseous out-put stream from a coal to acetylene conversion process. The gaseous out-put stream is initially treated in an acid gas removal stage by absorbing HCN and H_2S in an organic solvent such as N-methyl pyrrolidone and scrubbing with a caustic agent such as NaOH to remove CO_2. In a second stage, the gaseous out-put stream is scrubbed with the organic solvent to provide a sweet gas treatment and separate pure acetylene as a product. In a third stage, the gases deriving from second stage are first hydrogenated, then desulfurized and then methanated. The out-put stream from the

third stage is recycled to the coal to acetylene conversion process. In a fourth stage, the organic solvent from said second stage is refined and recycled to the first stage and/or second stage.

The disclosures of all above-mentioned reference documents are incorporated herein in entirety by references.

In the above introduction and description of prior art, it is apparent to those skilled in the prior art that single-stage reactor for cracking or decomposing carbonaceous material exits a lot of defects needed to urgently addressed. For example, the height of the single-stage reactor could not be lengthened freely as it needs to increase or optimize the reaction time of the carbonaceous material powder according to the reaction temperature as requested by the maximum yield of cracked products, for example acetylene. The rapid drop of hot stream temperature along the single-stage reactor longitudinal direction results in that the reacting jet flow must be quenched to maximize the cracked products yield in a relatively short moving distance and hence the carbonaceous material to cracked products conversion is greatly limited. Meantime as previously stated, serious energy waste and over-high temperature distribution in neighbor of reactor inner wall also are big technical problems not to allow being ignored.

The following description represents a new understanding of the reactor and process directed specifically to thermal decomposition of solid carbonaceous materials having volatile content to maximize the yield of some specific cracked products from the solid carbonaceous matter. At the same time, the necessary process parameters are further provided for heating the solid carbonaceous particles as fast as possible to decompose the particle releasing volatiles as fast as possible so as to avoid the char forming by secondary reactions of these volatiles in the solid carbonaceous particles.

On the basis of above analysis, via numerous attempts and experiments, the inventors finally invented a new apparatus for cracking or decomposing solid carbonaceous

material almost solving all of above mentioned defects, for example obtaining good contact efficiency between carbonaceous material fine powder and plasma stream, i.e. multi-stage plasma reactor which of working mechanism is much different from fluid-powder mixing concept in present single-stage plasma reactor.

SUMMARY OF THE INVENTION

In accordance with the first aspect of the present invention, there is provided a multi-stage plasma reactor for cracking carbonaceous material comprising:

first stage of reaction tube mainly for mixing of the carbonaceous material, carrier gas with first heating gas and pyrolysis of the carbonaceous material;

second stage-N stage of reaction tube mainly for gas phase reaction of volatiles resulted from the pyrolysis, wherein N is integer of more than or equal to 2;

at least one inlet of the carbonaceous material and carrier gas as feedstock located on top of the first stage of reaction tube;

at least one inlet of first heating gas located on side surface of the first stage of reaction tube;

at least one inlet of second-N heating gases located on side surface of the second stage-N stage of reaction tube respectively, wherein said second-N heating gases are plasma high temperature gases;

at least one inlet of quench media for quenching or freezing of reaction products;

at least one outlet of quenched products and gases located on bottom or lower portion of last stage of reaction tube;

wherein the carbonaceous material goes downstream from the top of the first stage of reaction tube to finally reach the bottom or lower portion of last stage of reaction tube

while completion of the pyrolysis, gas phase reaction and quench.

In the above mentioned multi-stage plasma reactor, it is preferred that the operation temperature in the first stage of reaction tube makes sure that the temperature of the carbonaceous material entering therein reaches 650-1250℃ while the operation temperatures in the second stage-N stage of reaction tube makes sure that the temperature of the gas phase reaction occurring therein reaches 1500-2900℃. The first heating gas preferably is H_2, N_2, methane, inert gas and/or plasma gases of H_2, N_2, methane, and/or inert gases while the second-N heating gases preferably are plasma high temperature gases of hydrogen, nitrogen, methane, and/or inert gas. On the other hand, the quench media entering said reactor should make sure that the reaction products therein are preferably quenched to below temperature of 527℃ before exiting said reactor.

It is also preferred that time for the gas phase reaction of volatiles resulted from the pyrolysis occurring in the second stage-N stage of reaction tube is 0.4 – 4.0 millisecond while total time for the pyrolysis, gas phase reaction and quench occurring in said reactor is less than 50 millisecond, for example 30 or 40 milliseconds. Generally, said quench media could include water, steam, propane, aromatics, inert gas, any types of carbonaceous material, and/or mixture thereof while said carrier gas could be selected from group consisting of hydrogen, methane, nitrogen, gaseous carbonaceous material, inert gas and/or mixture thereof. Especially, said inert gas could be argon.

The cross section of said reaction tube could optionally or preferably be round, square, elliptic, polygonal or any regular shape else, more preferably, cross section area of said second stage-N stage of reaction tube is 1-3 times of that of the first stage of reaction tube. Furthermore, the amount of the inlets of the carbonaceous material and carrier gas preferably is 1-100; amount of the inlets of the first-N heating gases preferably is 2-32 while amount of the inlets of quench media preferably is 8-100. Particularly preferably, the above said inlets are symmetrically and oppositely

arranged in the horizontal direction.

By broad definition, said the carbonaceous material could be solid, liquid and/or gaseous material, particularly, the carbonaceous material could further be selected from group consisting of coal, coal tar, coal direct liquefaction residue, heavy crude residuum, char, petroleum coke, tar sand, shale oil, carbonaceous industrial wastes or tailings, biomass, synthetic plastic, synthetic polymer, spent tire, municipal solid waste, bitumen, and mixture thereof.

Generally, said plasma high temperature gases and the plasma gases of H_2, N_2, methane, and/or inert gases could be generated by a plasma generator with power input of 10 kW - 20 MW.

To obtain better quench effect, angle formed by said inlets of first-N heating gases and the inlets of the quench media preferably is in ranged of from -45^o to $+45^o$ relative to the horizontal level. In the same way, to obtain better mixing effect and optimal residence time, both opposite or non-direct opposite the inlets of first-N heating gases, on the same horizontal level, preferably form an angle along with vertical direction, more preferably, angle, formed along with vertical direction, between both opposite or non-direct opposite the inlets of first heating gas on the same horizontal level could be larger than that, formed along with vertical direction, between both opposite or non-direct opposite the inlets of second-N heating gases on the same horizontal level.

It should be well understood to those skilled in the prior art that angles, formed along with vertical direction, between both opposite or non-direct opposite the inlets of second-N heating gases on the same horizontal level could be same or different.

In accordance with the second aspect of the present invention, there is provided a process for cracking carbonaceous material by using the above multi-stage plasma reactor comprising:

a) introducing the carbonaceous material by aid of the carrier gas via said inlets of the

carbonaceous material and carrier gas as feedstock into top of the first stage of reaction tube;

b) introducing the first heating gas jet into said first stage of reaction tube via the inlets of the first heating gas, wherein the carbonaceous material and carrier gas are forced to be mixed with the first heating gas, and the carbonaceous material then is pyrolyzed by thermal effect of the first heating gas jet;

c) Introducing the second-N heating gases into the second- N stages of reaction tube via the inlets of the second-N heating gases respectively, wherein the gas phase reaction of volatiles resulted from pyrolysis occurs therein, and optionally, the carbonaceous material continue to be pyrolyzed by thermal effect of the second-N heating gas jets, so as to produce cracked and/or pyrolyzed products;

d) introducing the quench media into said reactor via said quench media inlets so as to quench or freeze said cracked and/or pyrolyzed products;

e) withdrawing the cracked and/or pyrolyzed products, gases, and/or residuum of pyrolyzed carbonaceous material out of said reactor via said outlets of quenched products and gases.

In accordance with the third aspect of the present invention, there is provided a process for producing acetylene by using the above multi-stage plasma reactor comprising:

a) introducing the carbonaceous material by aid of the carrier gas via said inlets of the carbonaceous material and carrier gas as feedstock into top of the first stage of reaction tube;

b) introducing the first heating gas jet into said first stage of reaction tube via the inlets of the first heating gas, wherein the carbonaceous material and carrier gas are forced to be mixed with the first heating gas, and the carbonaceous material then is

pyrolyzed by thermal effect of the first heating gas jet;

c) Introducing the second-N heating gases into the second- N stages of reaction tube via the inlets of the second-N heating gases respectively, wherein the gas phase reaction of volatiles resulted from pyrolysis occurs therein, and optionally, the carbonaceous material continue to be pyrolyzed by thermal effect of the second-N heating gas jets, so as to produce cracked and/or pyrolyzed products;

d) introducing the quench media into said reactor via said quench media inlets so as to quench or freeze said cracked and/or pyrolyzed products;

e) withdrawing the cracked and/or pyrolyzed products, gases, and/or residuum of pyrolyzed carbonaceous material out of said reactor via said outlets of quenched products and gases.

f) separating acetylene from the cracked and/or pyrolyzed products and gases.

In the process according to second or third aspect of the present invention, the cracked and/or pyrolyzed products generally include acetylene, carbon monoxide, methane, ethylene, and char etc. At the same time, to obtain optimized dispersion effect and heating rate of the carbonaceous material, the carbonaceous material preferably has average particle diameter in range of 10-300 um, and temperature of the carbonaceous material before entering said reactor preferably is in range of 20-300℃ while volume ratio of the carbonaceous material to carrier gas preferably ranges from 10/90 – 90/10.

The operation pressure of the said reactor could generally be in range of from negative pressure to positive pressure. The heating rate of the carbonaceous material in the first stage of reaction tube preferably is more than 10^4 K/Second. The cracked products also are preferably quenched or frozen within 4 milliseconds after their formulation. Temperatures, flow rates, and/or species of said second-N heating gases could be same or different.

BRIEF DESCRIPTION OF DRAWINGS

FIG.1 is a representative schematic view of two-stage plasma reactor according to the present invention;

FIG.2 is a representative schematic view of three-stage plasma reactor according to the present invention;

SPECIFIC MODES OF IMPLEMENTING THE INVENTION

The present invention will be further illustrated in detail by the following description where references are made to the appended drawings, in which, the corresponding or equivalent parts or elements as shown in the drawings are represented by the same reference number.

Generally, the reaction of the volatiles released from pyrolysis of carbonaceous material such as bituminous coal play an essential and important part in the cracked products production. Because the carbonaceous material undergoes extremely fast reaction with highly reactive gases, for example plasma high temperature gases, and such reaction is required to be terminated instantaneously, so such pyrolysis and reaction could not be described or calculated by conventional processes. Broadly speaking, the cracked products distribution depends on the types of the carbonaceous materials and operation conditions employed, if the reaction and/or residence time is only a few milliseconds it is impossible to have sufficient time to reach thermal-dynamic equilibrium, therefore soot caused by secondary reaction of volatiles derived from the carbonaceous material could not form, in measurable amount.

For the solid carbonaceous material, the thermal transfer and pyrolysis of the carbonaceous material, homogeneous solid-gas phase reactions and homogeneous gas phase reactions all contribute to the rate of cracked products formation i.e. their yield. In fact, one of the main purposes of the present invention is to maximize the yield of some specific cracked products, for example acetylene, thereby it is key point to

understand and determine nature, mechanism and operation conditions of above pyrolysis and reactions for completion of the present invention.

It is attested by numerous measures and observations that pyrolysis temperature of the carbonaceous material, especially for solid carbonaceous material, is preferably in the range of 650-1250℃, for example 680-1100℃, more preferably 700 - 930℃, particularly preferably 750-900℃, for example 850℃ while the gas phase reaction temperature of the volatiles derived from the carbonaceous material preferably is in the range of 1500-2900℃, more preferably 1500-2500℃, particularly preferably 1500 - 2000℃, for example 1750℃ or 1850℃. The above temperatures decide the preferable operation temperature of first stage of said reaction tube and the preferable operation temperature of second-N stages of said reaction tube because the pyrolysis of the carbonaceous material mainly take place in the first stage where more than 60 % maximum production amount of volatiles generally is generated while the above gas phase reactions of the above volatiles mainly occur in the second-N stages of said reaction tube.

To obtain desired fast and completed conversion as possible, the above gas phase reaction time of the said volatiles preferably is less than 4 millisecond, for example 2 millisecond, more preferably less than 1millisecond, particularly preferably less than 0.4 millisecond, for example less 0.3 or 0.2 milliseconds. Such reaction time could guarantee to obtain high yield of the cracked products.

In general, there are several practical ways to enhance yield of cracked products, especially acetylene in following:

Firstly, the yield of some specific cracked products, for example acetylene is highly governed by the very fast reactions between primary volatiles and high reactive short-lived plasma species in the plasma high temperature gases, for instance plasma hydrogen and/or inert gases including helium. In consequence, the yield of cracked products is relatively high if the high concentration of or large amount of volatiles is

released by very rapid pyrolysis of the carbonaceous material, this can be achieved by reasonable selection of very fine particle size distribution of carbonaceous material, or application of type of the carbonaceous material in low rank and simultaneously with low oxygen content in consideration of the oxygen of the volatiles could be converted into carbon monoxide at the expense of acetylene.

Secondly, optimal operation conditions the pyrolysis of the carbonaceous material are selected to obtain maximum amount of volatiles. Therefore suitable pyrolysis time, pressure and/or temperature are key points to maximize production of said volatiles.

Thirdly, the above volatiles is brought into contact with as much of the highly reactive plasma species as possible, such practice could increase surface for reaction and enhance reaction conversion.

Fourthly, the reaction temperature at which the volatiles reacts with the highly reactive plasma species is another important factor affecting the yield of cracked products, generally, yield of the cracked products increases with increase of temperature of the above gas phase reaction, but over high reaction temperature will cause formation of soot and hydrogen in measurable amount.

It is apparent from the above analysis to those skilled in the art that pyrolysis of the carbonaceous material and gas phase reaction of the primary volatiles with high reactive plasma species both are the most important processes to production of cracked products. However, optimal process parameters or operation conditions of pyrolysis usually is not the same that of the above gas phase reactions, if like as design of the structure of single stage plasma reactor present in the prior art, i.e. pyrolysis and gas phase reaction take places in the same space or region, process parameters or operation conditions of the pyrolysis and gas phase reactions could not reach excellent balance and not be optimized.

With respect to the above fatal defect, the present inventor proposes and invents a

novel structure of multi-stage plasma reactor via numerous attempts and experiments, the invented multi-stage plasma reactor smartly causes occurrence of the above pyrolysis and gas phase reactions in different spaces or regions so as to simultaneously reach optimized process parameters or operation conditions of the pyrolysis and gas phase reactions.

In detail, as shown in FIGs. 1-2, said multi-stage plasma reactor comprises:

first stage of reaction tube mainly for mixing of the carbonaceous material, carrier gas with first heating gas and pyrolysis of the carbonaceous material;

second stage-N stage of reaction tube mainly for gas phase reaction of volatiles resulted from the pyrolysis, wherein N is integer of more than or equal to 2;

at least one inlet of the carbonaceous material and carrier gas as feedstock located on top of the first stage of reaction tube;

at least one inlet of first heating gas located on side surface of the first stage of reaction tube;

at least one inlet of second-N heating gases located on side surface of the second stage-N stage of reaction tube respectively, wherein said second-N heating gases are plasma high temperature gases;

at least one inlet of quench media for quenching or freezing of reaction products;

at least one outlet of quenched products and gases located on bottom or lower portion of last stage of reaction tube;

wherein the carbonaceous material goes downstream from the top of the first stage of reaction tube to finally reach the bottom or lower portion of last stage of reaction tube while completion of the pyrolysis, gas phase reaction and quench.

The structure design of the above multi-stage plasma reactor causes possibility that the process parameters and operation conditions of the pyrolysis and gas phase reaction are independently managed or selected respectively, and could simultaneously be optimized, that could not be reached by design of the structure of prior single stage plasma reactor in any way.

In the above multi-stage plasma reactor, because the temperature of pyrolysis is much less than temperature of gas phase reaction, the operation temperature of the first stage of the reaction tube, where the pyrolysis mainly takes place could be less than that of second-N stage of the reaction tube, where the gas phase reaction of primary volatiles with highly reactive plasma species mainly occurs, therefore the first heating gas for pyrolysis could be H_2, N_2, methane, inert gas, and/or plasma gases of H_2, N_2, methane, and/or inert gases while the second-N heating gases could be plasma high temperature gases of hydrogen, nitrogen , methane, and/or inert gas.

In order to prevent cracked products obtained in the gas phase reaction, for example acetylene from decomposing or occurrence of second reaction for finally forming low valuable soot and hydrogen, the generated cracked products must be quenched instantaneously before exiting said reactor. Generally, the cracked products preferably are quenched within 4 milliseconds, for example 2 milliseconds after their formulation to below temperature of 650℃, preferably below 600℃, particularly preferably below 527℃. Said quench media could preferably include water, steam, propane, aromatics, inert gas, any types of carbonaceous material and/or mixture thereof.

The pressure of said reactor system could be in range of from negative pressure to positive pressure, for example 70-200 KPa, preferably 100-150 KPa, more preferably 110-140 KPa. The length of reactor and feedstock flow rate typically depend on the residence time of feedstock in the every stage of reaction tube and reactions time. More typically, the total time for the pyrolysis, gas phase reaction and quench occurring in said reactor is preferably less than 50 milliseconds.

To obtain excellent transportation efficiency of the carbonaceous material in the very fine particles or well dispersion form and/or realize well mixing or intimate contact of the carbonaceous material with heating gases, carrier gas for transposition of said carbonaceous material is generally required, and could be selected from group consisting of hydrogen, methane, nitrogen, gaseous carbonaceous material, inert gas and/or mixture thereof. The exemplary examples of inert gases are for example argon and/or helium.

The cross section of said reaction tube could be in any shape, for example round, square, elliptic, polygonal or any regular shape else. But to prevent inner surface of the reaction tube wall from apparently coking, cross section area of said second stage-N stage of reaction tube preferably is 1-3 times of that of the first stage of reaction tube. Such design prevents feedstock or cracked products from direct flushing the above inner surface and forming or accumulating coking thereon.

In the same way, to uniformly distribute or disperse feedstock, heating gases, cracked products and /or quench media in the inner space of said reactor, it is preferred that amount of the inlets of the carbonaceous material and carrier gas is 1-100, amount of the inlets of the first-N heating gases is 2-32 while amount of the inlets of quench media is 8-100, furthermore above said inlets more preferably are symmetrically and oppositely arranged in the horizontal direction.

The carbonaceous material useful in the multi-stage plasma reactor according to present invention could be solid, liquid and/or gaseous material, but preferably solid carbonaceous material, for example is selected from group consisting of coal, coal tar, coal direct liquefaction residue, heavy crude residuum, char, petroleum coke, tar sand, shale oil, carbonaceous industrial wastes or tailings, biomass, synthetic plastic, synthetic polymer, spent tire, municipal solid waste, bitumen, and/or mixture thereof.

In the multi-stage plasma reactor according to the present invention, the used plasma high temperature gases and/or the plasma gases of H_2, N_2, methane, and/or inert gases

could be generated by a plasma generator with power input of 10 kW -20 MW. Detailed information about the above-mentioned plasma generator could be took from or are referred to the aforesaid reference documents, for example US4358629, CN1562922A or CN 101742808A, the detailed description regarding it is herein omitted for economic presentation.

Because the time of pyrolysis of carbonaceous material, which mainly takes place in the first stage of the reaction tube, usually is more than that of gas phase reaction of primary volatiles with highly reactive but very short-lived plasma species contained in the plasma high temperature gases, which mainly occurs in second –N stage of the reaction tube, therefore the residence time of feedstock in the first stage is much more than that of it in the second – N stage, to realize this arrangement, it is preferred that both opposite or non-direct opposite the inlets of first-N heating gases, on the same horizontal level, form an angle along with vertical direction, and the angle, formed along with vertical direction, between both opposite or non-direct opposite the inlets of first heating gas on the same horizontal level is larger than that, formed along with vertical direction, between both opposite or non-direct opposite the inlets of second-N heating gases on the same horizontal level, of course, angles, formed along with vertical direction, between both opposite or non-direct opposite the inlets of second-N heating gases on the same horizontal level could be same or different.

To obtain optimal quench efficiency of cracked products, especially fresh acetylene so as to maximize their yield, it is also preferred that angle formed by said inlets of first-N heating gases and the inlets of the quench media is in ranged of from -45^{o} to +45^{o} relative to the horizontal level.

The above novel structure design of multi-stage plasma reactor according to the present invention has following advantages and characteristics:

Firstly, the height of the multi-stage plasma reactor could be lengthened freely as it needs to increase or optimize the residence or reaction time of the carbonaceous

material powder according to the reaction temperature as requested by the maximum yield of cracked products, for example acetylene.

Secondly, the slow or lax drop, or even a little increase of hot stream temperature along the multi-stage plasma reactor longitudinal direction results in that the reacting jet flow could be quenched to maximize the cracked products yield in a relatively long moving distance and hence the carbonaceous material to cracked products conversion is greatly enhanced.

Thirdly, serious energy waste and over-high temperature distribution in neighbor of reactor inner wall could also be avoided because heating is done by stage by stage, or step by step, as a result, over concentration of thermal emission would not occur.

The multi-stage plasma reactor according to the present invention could be applied to produce cracked products derived from variety of carbonaceous material; typical process is described in following:

a) introducing the carbonaceous material by aid of the carrier gas via said inlets of the carbonaceous material and carrier gas as feedstock into top of the first stage of reaction tube;

b) introducing the first heating gas jet into said first stage of reaction tube via the inlets of the first heating gas, wherein the carbonaceous material and carrier gas are forced to be mixed with the first heating gas, and the carbonaceous material then is pyrolyzed by thermal effect of the first heating gas jet;

c) Introducing the second-N heating gases into the second- N stages of reaction tube via the inlets of the second-N heating gases respectively, wherein the gas phase reaction of volatiles resulted from pyrolysis occurs therein, and optionally, the carbonaceous material continue to be pyrolyzed by thermal effect of the second-N heating gas jets, so as to produce cracked and/or pyrolyzed products;

d) introducing the quench media into said reactor via said quench media inlets so as to quench or freeze said cracked and/or pyrolyzed products;

e) withdrawing the cracked and/or pyrolyzed products, gases, and/or residuum of pyrolyzed carbonaceous material out of said reactor via said outlets of quenched products and gases.

In general, the cracked products from the carbonaceous material is a mixture including acetylene, carbon monoxide, methane, ethylene, hydrogen, and char etc. if some specific cracked products, for example acetylene are intended to be obtained, such cracked products mixture is required to be separated so as to get essentially pure cracked product. For example, the aforesaid reference documents - US 4367363 disclosed such separation method in which the pure acetylene was separated from the above cracked products mixture. The detailed description about it is omitted herein for economic presentation.

In order to obtain optimal efficiency of pyrolysis and cracking of the carbonaceous material, in addition to the structure design of the multi-stage plasma reactor, the physical and chemical properties of feedstock should be further considered or selected so as to maximize the cracked products yield. In general, average particle diameter of the carbonaceous material preferably is in range of 10 - 300 um while temperature of the carbonaceous material before entering said reactor preferably is in range of 20 - 300℃. The volume ratio of the carbonaceous material to carrier gas generally ranges from 10/90 - 90/10, preferably 20/80-80/20, more preferably 30/70-70/30, particularly preferably 40/60-60/40, for example 50/50.

Finally, it should be understood that the heating rate of the carbonaceous material in the first stage of reaction tube preferably is more than 10^4 K/Second; and the temperatures, flow rates and/or species of said second-N heating gases could be same or different for operation flexibility and various operation requirements in different circumstances.

EXAMPLE

Example 1

A two-stage plasma reactor for cracking coal, which of schematic view was shown in Figure 1, was used to convert coal into acetylene and other chemicals. The two-stage plasma reactor is capable of operating by aid of plasma generator with power input ranging 10 kW - 20 MW. As shown in Figure 1, the reactor consisted of two-stage flat channel (i.e. reaction tube), three coal powder inlets at top, two first heating gas inlets, two second heating gas inlets, two quench media inlets and a cracked products outlet. The reactor wall was constructed of copper near the four heating gas inlets and steel at other regions while the reactor wall was cooled by water cycling at high velocity in the annulus gap between the wall and its external jacket.

The high volatile bituminous coal was grinded into coal powder fine particles with a particle size distribution (PSD) of 72% by weight <106 micrometer and 100 by weight <150 micrometer. As a feedstock, the coal powder at 300° K was injected into top of the first stage of reaction tube through the coal powder inlets along with hydrogen as a carrier gas. The coal powder contained about 40% by weight of volatiles by proximate analysis, accounted by dry and ash free base while by the ultimate analyses, the coal powder had elemental composition, accounted by dry bases (dried for 2 hours at 110℃), as shown in below table 1:

Table 1

C w%	H w%	O w%	N w%	S w%	H_2O w%	Ash w%
79.2	5.5	6.3	1.6	1.1	2.3	4.0

The reactor ran at the following operation conditions: system pressure of 115 kPa, input power of plasma generator of 60 kW, coal flow rate of 30 kg/h, and hydrogen flow rate 4.2 kg/h (where 4.0 kg/h as heating gas and 0.2 kg/h as the carrier gas). The hydrogen as heating gas was heated to plasma hydrogen at the temperature of about 2600 K so as for heat conversion factor to reach about 82% and was then injected into

the first stage and second stage of reaction tube through the two first heating gas inlets and two second heating gas inlets equally. Water was injected into inside of the reactor through two quench media inlets near the outlet of cracked products to instantaneously quench or freeze the formed products stream. The total residence time of coal powder in the reactor was approximately 40 milliseconds. It was estimated by heat transfer calculation and energy balance that the reactor had about 76% energy efficiency, i.e., 76% of the power input was intaken by the products stream and the water cooling wall of the plasma generators and plasma reactor in which the heat loss cause by the reactor was about 4.8 kW.

The reactor output products stream, formed under the above operation conditions, had the acetylene yield and energy consumption, as shown in below table 2:

Table 2

Acetylene/100 kg coal	SER
19.0 kg	10.5 kWh/kg-C_2H_2

In the above table 2, SER is referred to the gross Specific Energy Requirement based on the power delivered at the electrodes.

COMPARATIVE EXAMPLE 1

The experiment described in the example1 was repeated except that the two-stage plasma reactor was substituted by prior single-stage plasma reactor, in which two second heating gas inlets were deleted and the same amount of heating gas was injected into the single stage of reaction tube through two first heating gas inlets.

The performances of the two different types of reactors were listed in below table 3. It is apparent from the table 3 by comparison that performances of the two-stage plasma reactor according to the present invention were much better than that of prior single stage plasma reactor except that coal conversion rate of the two-stage plasma reactor

was a bit lower than that of the prior single-stage one.

Table 3

Example	Com-example 1	Example 1
The type of reactor	Single-stage reactor	Two-stage reactor
Acetylene yield, kg/100 kg coal	15.8	19.0
SER, kWh/kg-C_2H_2	12.7	10.5
Mass Fraction of C_2H_2 in Products Stream, wt %	26.2	31.7
Coal Conversion, %	46.2	46.0
Heat Flow Rate Loss from Reactor Wall, kW	5.5	4.8
Energy Efficiency %	73.2	76.1

In the above table 3, the meaning of SER was the same as that in the table 2; Energy Efficiency was referred to relative amount of heat intaken by products steam and cooling water compared to power input.

EXAMPLE 2

The experiment described in the example1 was repeated except that the two-stage plasma reactor was substituted by tri-stage plasma reactor, in which the tri-stage plasma reactor is capable of operating by aid of plasma generator with power input ranging 10 kW -20 MW. As shown in Figure 2, the reactor consisted of three-stage flat channel (i.e. reaction tube), three coal powder inlets at top, two first heating gas inlets, two second heating gas inlets, two third heating gas inlets, two quench media inlets and a cracked products outlet. The reactor wall was constructed of copper near the six heating gas inlets and steel at other regions while the reactor wall was cooled by water cycling at high velocity in the annulus gap between the reactor wall and its external jacket. Coal powder used in the present example 2 was the same as that used in the example 1, and was grinded into fine powder with a particle size distribution (PSD) of 80% by weight <100 micrometer and 100 by weight <120 micrometer.

The reactor ran at the following operation conditions: system pressure of 125 kPa, input power of plasma generator of 80 kW, coal flow rate of 40 kg/h, and hydrogen

flow rate 5.25 kg/h (where 5.0 kg/h as heating gas and 0.25 kg/h as the carrier gas). The hydrogen as heating gas was heated to plasma hydrogen at the temperature of about 2800 K so as for heat conversion factor to reach about 84% and was then equally injected into the first stage, second stage and third stage of reaction tube through the two first heating gas inlets, two second heating gas inlets, and two third heating gas inlets. Water was injected into inside of the reactor through two quench media inlets near the outlet of cracked products to instantaneously quench or freeze the formed products stream. The total residence time of coal powder in the reactor was approximately 35 milliseconds. It was estimated by heat transfer calculation and energy balance that the reactor had about 78.2% energy efficiency, i.e., 78.2% of the power input was intaken by the products stream and water cooling wall of the plasma generators and plasma reactor in which the heat loss cause by the reactor was about 5.0 kW.

The reactor output products stream, formed under the above operation conditions, had the acetylene yield and energy consumption, as shown in below table 4:

Table 4

Acetylene/100 kg coal	SER
19.5 kg	10.3 kWh/kg-C_2H_2

In the above table 4, SER is referred to the gross Specific Energy Requirement based on the power delivered at the electrodes.

COMPARATIVE EXAMPLE 2

The experiment described in the example 2 was repeated except that the tri-stage plasma reactor was substituted by prior single-stage plasma reactor, in which two second heating gas inlets and two third heating gas inlets were deleted and the same amount of heating gas was injected into the single stage of reaction tube through two first heating gas inlets.

The performances of the two different types of reactors were listed in below table 5. It is apparent from table 5 by comparison that performances of the tri-stage plasma reactor according to the present invention were much better than that of prior single stage plasma reactor, even better than that of the two-stage plasma reactor described in the example 1 except that coal conversion rate of the tri-stage plasma reactor was a bit lower than that of the prior single-stage one.

Table 5

Example	Comparative example 2	Example 2
The type of reactor	Single-stage reactor	Tri-stage reactor
Acetylene yield, kg/100 kg coal	15.5	19.5
SER, kWh/kg-C_2H_2	12.9	10.3
Mass Fraction of C_2H_2 in Product Stream, wt %	25.7	32.3
Coal Conversion, %	47.3	47.2
Heat Flow Rate Loss from Reactor Wall, kW	6.1	5.0
Energy Efficiency %	74.5	78.2

In the above table 5, the meaning of SER was the same as that in the table 4; Energy Efficiency was referred to relative amount of heat intaken by products steam and cooling water compared to power input.

The terms and expressions which have been employed in this specification are used only as terms and expressions of description and not of limitations, and there is no intention in the use of such terms and expressions to exclude any equivalents of the features shown and described or portions thereof.

Although a few embodiments of the present invention have been shown and described, the present invention is not limited to the described embodiments. Instead, it would be appreciated by those skilled in the art that any changes and modification may be made to these embodiments without departing from the principles and spirit of the invention, the scope of which is defined by the appended claims and their equivalents

Claims

1、A multi-stage plasma reactor for cracking carbonaceous material comprising:

first stage of reaction tube mainly for mixing of the carbonaceous material, carrier gas with first heating gas and pyrolysis of the carbonaceous material;

second stage-N stage of reaction tube mainly for gas phase reaction of volatiles resulted from the pyrolysis, wherein N is integer of more than or equal to 2;

at least one inlet of the carbonaceous material and carrier gas as feedstock located on top of the first stage of reaction tube;

at least one inlet of first heating gas located on side surface of the first stage of reaction tube;

at least one inlet of second-N heating gases located on side surface of the second stage-N stage of reaction tube respectively, wherein said second-N heating gases are plasma high temperature gases;

at least one inlet of quench media for quenching or freezing of reaction products;

at least one outlet of quenched products and gases located on bottom or lower portion of last stage of reaction tube;

wherein the carbonaceous material goes downstream from the top of the first stage of reaction tube to finally reach the bottom or lower portion of last stage of reaction tube while completion of the pyrolysis, gas phase reaction and quench.

2、The multi-stage plasma reactor according to claim 1, in which the operation temperature in the first stage of reaction tube makes sure that the temperature of the carbonaceous material entering therein reaches 650-1250℃ while the operation temperatures in the second stage-N stage of reaction tube makes sure that the

temperature of the gas phase reaction occurring therein reaches 1500-2900℃.

3、The multi-stage plasma reactor according to claim 1, in which the first heating gas is H_2, N_2, methane, inert gas and/or plasma gases of H_2, N_2, methane, and/or inert gases while the second-N heating gases are plasma high temperature gases of hydrogen, nitrogen, methane, and/or inert gas.

4、The multi-stage plasma reactor according to claim 1, in which the quench media entering said reactor makes sure that the reaction products therein are quenched to below temperature of 527℃ before exiting said reactor.

5、The multi-stage plasma reactor according to claim 1, in which time for the gas phase reaction of volatiles resulted from the pyrolysis occurring in the second stage-N stage of reaction tube is 0.4 – 4 millisecond.

6、The multi-stage plasma reactor according to claim 1, in which total time for the pyrolysis, gas phase reaction and quench occurring in said reactor is less than 50 millisecond.

7、The multi-stage plasma reactor according to claim 1, in which said quench media includes water, steam, propane, aromatics, inert gas, any types of carbonaceous material and/or mixture thereof.

8、The multi-stage plasma reactor according to claims 1, in which said carrier gas is selected from group consisting of hydrogen, methane, nitrogen, gaseous carbonaceous material, inert gas and/or mixture thereof.

9、The multi-stage plasma reactor according to any one of aforesaid claim 3,7 and 8, in which said inert gas is argon.

10、The multi-stage plasma reactor according to claim 1, in which cross section of said reaction tube is round, square, elliptic, polygonal or any regular shape else.

11、The multi-stage plasma reactor according to claim 1, in which cross section area of said second stage-N stage of reaction tube is 1-3 times of that of the first stage of reaction tube.

12、The multi-stage plasma reactor according to claim 1, in which amount of the inlets of the carbonaceous material and carrier gas is 1-100, amount of the inlets of the first-N heating gases is 2-32 while amount of the inlets of quench media is 8-100.

13、The multi-stage plasma reactor according to any one of aforesaid claims 1-8 and 10-12, in which said inlets are symmetrically and/or oppositely arranged in the horizontal direction.

14、The multi-stage plasma reactor according to any one of aforesaid claims 1-8 and 10-12, in which said the carbonaceous material is solid, liquid and/or gaseous material.

15、The multi-stage plasma reactor according to any one of aforesaid claims 1-8 and 10-12, in which the carbonaceous material further is selected from group consisting of coal, coal tar, coal direct liquefaction residue, heavy crude residuum, char, petroleum coke, tar sand, shale oil, carbonaceous industrial wastes or tailings, biomass, synthetic plastic, synthetic polymer, spent tire, municipal solid waste, bitumen, and/or mixture thereof.

16、The multi-stage plasma reactor according to any one of aforesaid claims 1-8 and 10-12, in which said plasma high temperature gases and the plasma gases of H_2, N_2, methane, and/or inert gases are generated by a plasma generator with power input of 10 kW - 20MW.

17、The multi-stage plasma reactor according to any one of aforesaid claims 1-8 and 10-12, in which angle formed by said inlets of first-N heating gases and the inlets of the quench media is in range of from -45^{o} to $+45^{o}$ relative to the horizontal level.

18、The multi-stage plasma reactor according to any one of aforesaid claims 1-8 and 10-12, in which both opposite or non-direct opposite the inlets of first-N heating gases, on the same horizontal level, form an angle along with vertical direction.

19、The multi-stage plasma reactor according to claim 18, in which angle, formed along with vertical direction, between both opposite or non-direct opposite the inlets of first heating gas on the same horizontal level is larger than that, formed along with vertical direction, between both opposite or non-direct opposite the inlets of second-N heating gases on the same horizontal level.

20、The multi-stage plasma reactor according to any one of aforesaid claims 1-8 and 10-12, in which angles, formed along with vertical direction, between both opposite or non-direct opposite the inlets of second-N heating gases on the same horizontal level are same or different.

21、A process for cracking carbonaceous material by using the multi-stage plasma reactor according to any one of aforesaid claims1-20 comprising:

a) introducing the carbonaceous material by aid of the carrier gas via said inlets of the carbonaceous material and carrier gas as feedstock into top of the first stage of reaction tube;

b) introducing the first heating gas jet into said first stage of reaction tube via the inlets of the first heating gas, wherein the carbonaceous material and carrier gas are forced to be mixed with the first heating gas, and the carbonaceous material then is pyrolyzed by thermal effect of the first heating gas jet;

c) Introducing the second-N heating gases into the second- N stages of reaction tube via the inlets of the second-N heating gases respectively, wherein the gas phase reaction of volatiles resulted from pyrolysis occurs therein, and optionally, the carbonaceous material continue to be pyrolyzed by thermal effect of the second-N heating gas jets, so as to produce cracked and/or pyrolyzed products;

d) introducing the quench media into said reactor via said quench media inlets so as to quench or freeze said cracked and/or pyrolyzed products;

e) withdrawing the cracked and/or pyrolyzed products, gases, and/or residuum of pyrolyzed carbonaceous material out of said reactor via said outlets of quenched products and gases.

22、A process for producing acetylene by using the multi-stage plasma reactor according to any one of aforesaid claims1-20 comprising:

a) introducing the carbonaceous material by aid of the carrier gas via said inlets of the carbonaceous material and carrier gas as feedstock into top of the first stage of reaction tube;

b) introducing the first heating gas jet into said first stage of reaction tube via the inlets of the first heating gas, wherein the carbonaceous material and carrier gas are forced to be mixed with the first heating gas, and the carbonaceous material then is pyrolyzed by thermal effect of the first heating gas jet;

c) Introducing the second-N heating gases into the second- N stages of reaction tube via the inlets of the second-N heating gases respectively, wherein the gas phase reaction of volatiles resulted from pyrolysis occurs therein, and optionally, the carbonaceous material continue to be pyrolyzed by thermal effect of the second-N heating gas jets, so as to produce cracked and/or pyrolyzed products;

d) introducing the quench media into said reactor via said quench media inlets so as to quench or freeze said cracked and/or pyrolyzed products;

e) withdrawing the cracked and/or pyrolyzed products, gases, and/or residuum of pyrolyzed carbonaceous material out of said reactor via said outlets of quenched products and gases;

f) separating acetylene from the cracked and/or pyrolyzed products and gases.

23、The process according to claim 21 or 22, in which the cracked and/or pyrolyzed products include acetylene, carbon monoxide, methane, ethylene, and char etc.

24、The process according to claim 21 or 22, in which average particle diameter of the carbonaceous material is in range of 10 - 300 um.

25、The process according to claim 21 or 22, in which temperature of the carbonaceous material before entering said reactor is in range of 20 - 300℃.

26、The process according to claim 21 or 22, in which volume ratio of the carbonaceous material to carrier gas ranges from 10/90 – 90/10.

27、The process according to claim 21 or 22, in which operation pressure of the said reactor is in range of from negative pressure to positive pressure.

28、The process according to claim 21 or 22, in which heating rate of the carbonaceous material in the first stage of reaction tube is more than 10^4 K/Second.

29、The process according to claim 21 or 22, in which the cracked products are quenched within 4 milliseconds after their formulation.

30、The process according to claim 21 or 22, in which temperatures, flow rates, and/or species of said second-N heating gases are same or different.

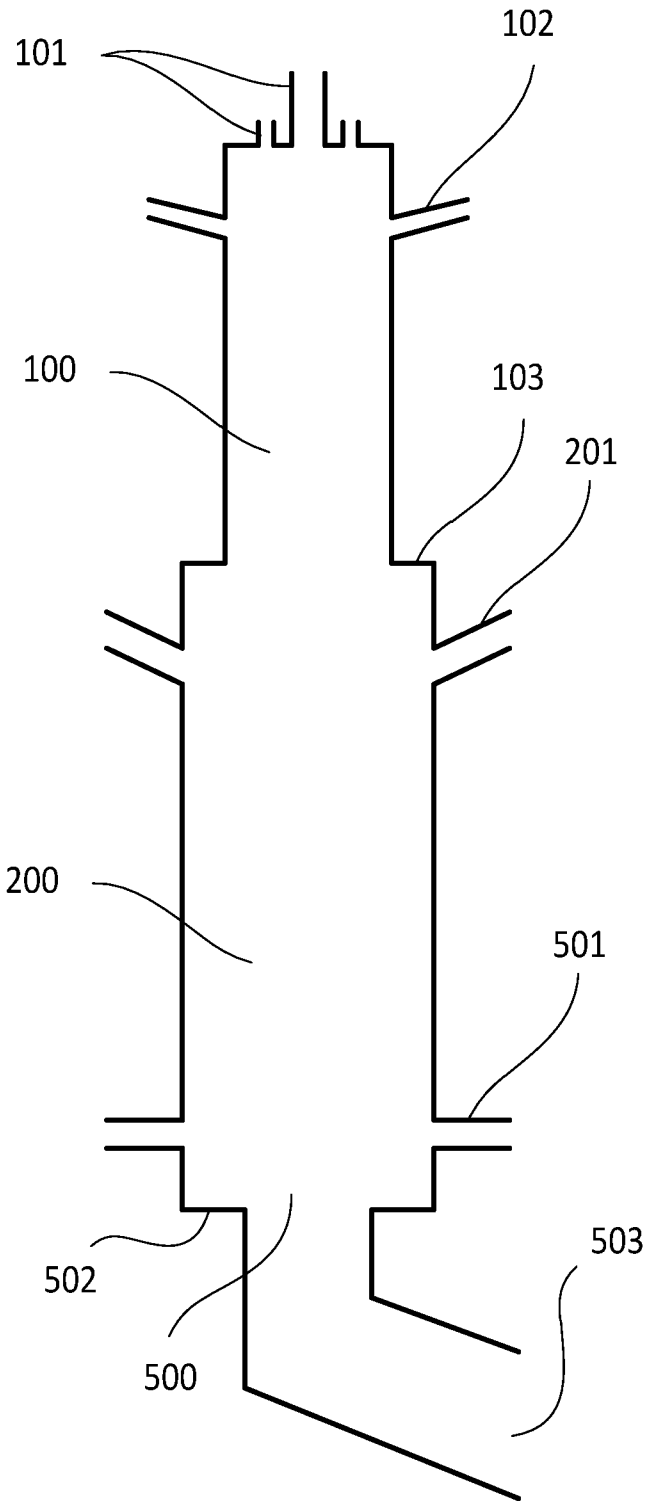


FIGURE 1

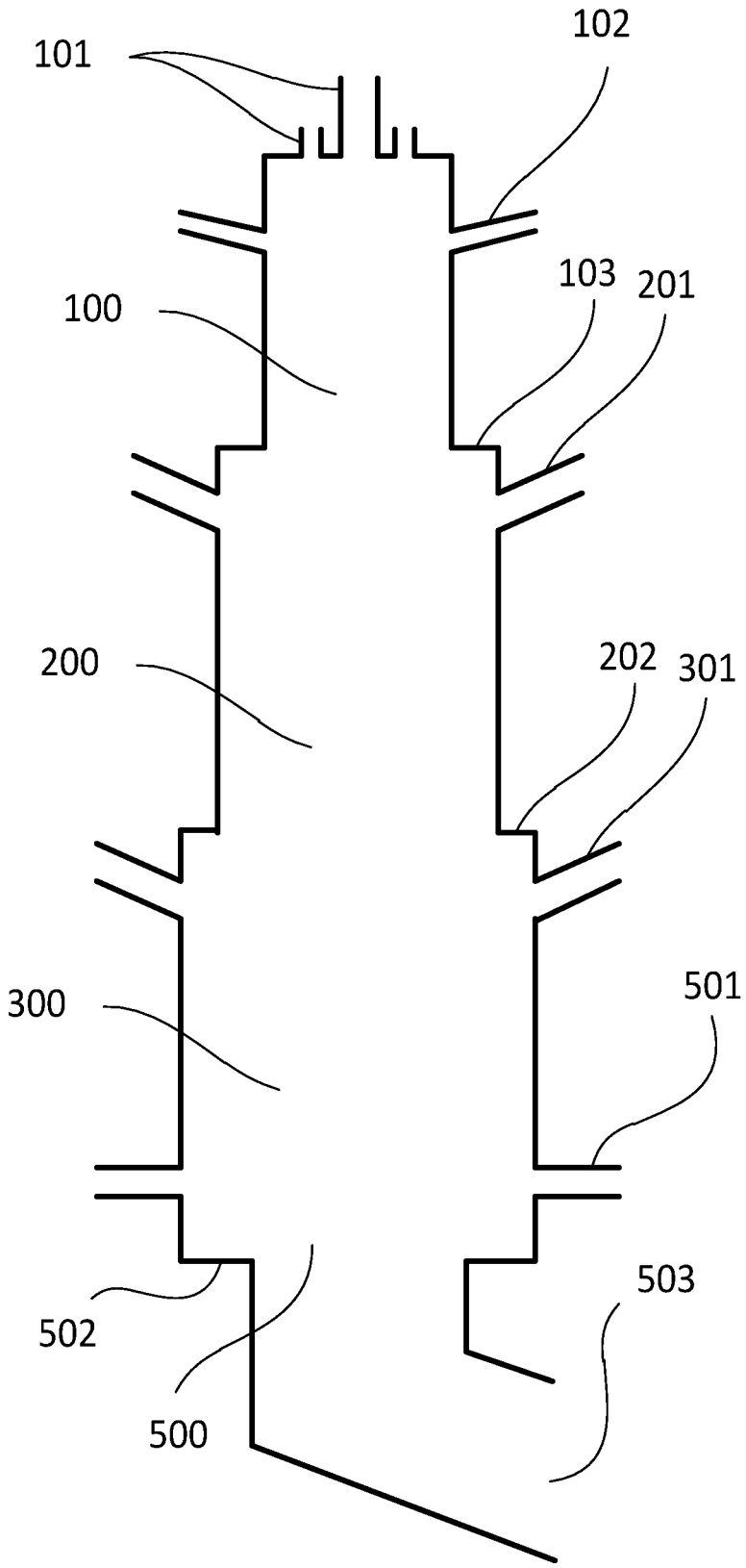


FIGURE 2

INTERNATIONAL SEARCH REPORT

International application No.
PCT/CN2012/079637

A. CLASSIFICATION OF SUBJECT MATTER

See extra sheet

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC: B01J19, C07C11, C07C4

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

CNPAT, CNKI, WPI, EPODOC: plasma, pyrolysis, crack+, heat+, thermal plasma, acetylene, ehtine, ethyne, quench+, freez+

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US3622493 A (CRUSCO FA) 23 Nov. 1971 (23.11.1971) column 1, lines 3-8; column 2, lines 12-16; column 3, lines 38-58; Fig. 1	1-30
A	CN101508623 A (TSINGHUA UNIVERSITY et al.) 19 Aug. 2009 (19.08.2009) Figs 1-3; Claims 1-9	1-30
A	CN1600758 A (TIAN Y) 30 Mar. 2005 (30.03.2005) Claims 1-3	1-30
A	QIU Jieshan et al. Pyrolysis of coal sunder plasma conditions to produce acetylene directly. Coal Chemical Engineering. 1994, 68(3). Pages 8-15, 27	1-30
A	US4358629 A (AVCO CORP) 09 Nov. 1982 (09.11.1982) Fig. 2	1-30

☒ Further documents are listed in the continuation of Box C. ☒ See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim (S) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
17 Oct. 2012 (17.10.2012)	**15 Nov. 2012 (15.11.2012)**
Name and mailing address of the ISA/CN The State Intellectual Property Office, the P.R.China 6 Xitucheng Rd., Jimen Bridge, Haidian District, Beijing, China 100088 Facsimile No. 86-10-62019451	Authorized officer Lei, Jun Telephone No. (86-10)62084747

Form PCT/ISA /210 (second sheet) (July 2009)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/CN2012/079637

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	CN1478858 A (UNIV TAIYUAN TECHNOLOG) 03 Mar. 2004 (03.03.2004) Fig. 1; Claims 1-9	1-30
A	CN1562922 A (UNIV TAIYUAN TECHNOLOG) 12 Jan. 2005 (12.01.2005) Fig. 1; Claims 1-7	1-30
A	CHEN Honggang, et al. Status quo and progress in the acetylene production from coal by plasma pyrolysis. Coal Conversion. Apr. 1996, vol. 19, No. 2 Pages 19-24	1-30

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/CN2012/079637

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		FR1560417 A	21.03.1969
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		DE6900459 U	14.12.1972
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US4358629 A	09.11.1982	GB2082198 A	03.03.1982
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INTERNATIONAL SEARCH REPORT

International application No.
PCT/CN2012/079637

Continuation of A: Classification of subject matter

B01J19/08 (2006.01) i
C07C11/24 (2006.01) i
C07C4/04 (2006.01) i