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(54) **Title:** PROCESS FOR THE PRODUCTION OF SYNTHESIS GAS BY MEANS OF A FLUID BED GASIFICATION REACTOR FED WITH CARBONACEOUS MATERIAL AND DEVICE SUITABLE FOR THE PURPOSE

(57) **Abstract:** A process for the production of synthesis gas which comprises : feeding a carbonaceous material having a particle-size lower than or equal to 50 mm, to a gasification reactor; feeding an oxidizing gas to the bottom of the reactor at a flow-rate which is such as to give the solid mass present in the reactor an aspect of a biphasic bed; filtering the solid particles entrained by the gas, in a cyclone filter positioned internally at the top of the reactor; discharging from the cyclone filter a gaseous stream essentially consisting of synthesis gas and unreacted oxidizing gas; and recycling the filtered solid particles, discharged at the base of the cyclone, directly to the biphasic bed, through an inner duct.

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PROCESS FOR THE PRODUCTION OF SYNTHESIS GAS BY MEANS OF A FLUID BED GASIFICATION REACTOR FED WITH CARBONACEOUS MATERIAL AND DEVICE SUITABLE FOR THE PURPOSE

DESCRIPTION

The present invention relates to a process for the production of synthesis gas by means of a fluid-bed gasification reactor fed with carbonaceous material.

More specifically, the present invention relates to a process for the production of synthesis gas using a fluid-bed gasification reactor fed with biomass.

Even more specifically, the present invention relates to this process and the device suitable for this purpose.

Processes for the conversion of carbonaceous materials, for example biomasses, into energy, can be divided into two main categories: thermochemical, of interest in the present invention, and biochemical. All conversion processes of biomasses into energy are based on the extraction of the heat content of the substance used or storage of this in a different energy carrier which can be subsequently used at a later moment.

In thermochemical processes, the action of heat allows solid fuel to be transformed into a gas mainly consisting of hydrogen and carbon monoxide which, after suitable purification, is suitable for direct use in modern energy conversion systems, for example gas turbines, for the production of electric energy or possible transformations into chemical products, for example hydrocarbons according to the Fischer-Tropsch synthesis. When the solid fuel is biomass, these processes are applied to products characterized by a humidity on collection lower than 30% and with a C/N ratio higher than 30.

In particular, gasification is a thermochemical transformation system of a solid reagent (for example biomasses or carbonaceous material) which is converted into synthesis gas (mainly H_2 and CO, with the presence of CO_2, CH_4, H_2O). This transformation takes place in an oxygen deficiency, with respect to the quantity necessary for the complete combustion. The oxidizing agents commonly used are air and oxygen possibly mixed with steam.

"Equivalence ratio" (ER) is the ratio between the oxygen used and stoichiometric oxygen, wherein ER is equal to 1 in the case of stoichiometric combustion, whereas the value ER = 0 corresponds to pyrolysis. Gasification generally requires an ER equal to 0.25-0.5.

The possible technologies for gasification, in particular in the presence of biomasses, are:

- fixed bed;
- fluid-bed (bubble, circulating, pressurized)
- entrained flow.

Fixed bed gasifiers are fed with relatively gross particle-sizes (with a maximum dimension even up to 100 mm). They are mainly used in medium-small-sized plants and are characterized by technological simplicity and economical running. The efficiencies however are not particularly high and they cannot tolerate excessively fine particles.

Fluid-bed gasifiers have a more efficient thermal exchange and greater control of the composition of the outgoing gas, but also a delicate fluid dynamics, with risks of packing and defluidization of the bed itself. The fluid bed technology is adequate for more reduced dimensions of the particles fed with respect to the fixed bed technology, preferably lower than 50 mm, but it tolerates a non-uniform distribution of the particles .

Entrained flow gasifiers allow a high power to be reached (hundreds of MW) but require very fine particles (=*100-400 μm). They are mainly used with liquid fuels or with coal. For biomasses, a previous torrefaction or alternative co-firing with coal is considered. The technology which is currently most promising, especially for the use of biomasses, is the fluid-bed technology.

Fluid-bed gasification has various basic principles which make it differ considerably from other types of gasification :

- the primary solid fuel (for example, biomass) is immersed in a bed of fluidized inert material, generally silica sand. In the case of fuels with a high ash content, the ash itself of the fuel can act as bed. There are also studies in which the inert bed is substituted with catalytically active material (for example, dolomite, olivine, etc.) as it favours reforming and water gas shift reactions between the gases gradually formed, modifying the composition of the gas phase directly in the gasifier itself and thus raising the H_2/CO ratio.

- the solid particles of biomass are kept in suspension (fluidization) by means of the flow of gaseous reagents injected under pressure from below (ascending stream) . The gaseous reagents consist of oxidizing agent (which can be air or oxygen) and possibly steam.

The turbulences typical of the fluid bed create a series of positive effects, for example:

- the mixing between solid particles and gas is excellent, the transfer of heat and matter between the flows involved in the gasification is high and efficient ;

- the reaction rates are higher with respect to fixed-bed reactors *(updraft* and *downdraft)* and consequently the residence time of the reagents is shorter, i.e. resulting in a lower volume of the reactor;

The fluid bed is also characterized by a uniform temperature distribution (isotherm) in the gasification zone, with temperatures typically within the range of 800÷900°C. This temperature range is obviously a compromise between the various requirements;

- resistance of the materials;

- the risk of sintering of the powders and slagging of the ashes, which dramatically increases if the temperature of the bed exceeds the melting point of the ashes. The agglomeration of the fine particles inside the bed should be avoided as it leads to a loss in the fluidization of the bed;

- the necessity of raising the conversion of the carbon and avoiding the formation of tar, for which an increase in the temperature of the bed would be desirable .

In conclusion, a fluid bed is considered as being a bed of solid particles situated in a duct at whose base gas is introduced with a certain surface rate (i.e. referring to the empty duct) defined as *Ug.* In addition to this parameter, there are a further two parameters which are fundamental in the fluid-dynamic analysis of the regimes of a fluid bed:

- the vacuum degree *(ε):* volumetric fraction occupied by the gas with respect to the overall volume used by the solid bed; and

- average slip rate *(Usl) :* difference between the effective average rates of the gas and solid, assumed positive if directed upwards.

Fluid-bed gasification envisages two basic types of reactor:

- Bubbling Fluidized Bed (BFB) ;
- Circulating Fluidized Bed (CFB) .

In the first type (BFB) , the reactor is composed of two sections having a different diameter, the upper part (larger diameter) allows a slowing-down of the gaseous stream and consequently the reflux of most of the solid particulate (gasification bed, non-combusted particles, powders, etc.); normally a cyclone is envisaged downstream, for separating any possible entrained fine products.

In the second configuration (CFB), the reactor has a larger quantity of entrained solids, which are separated in a cyclone situated downstream and reinserted into the gasifier through an outer pipe, complicating the plant design.

The Applicant has now found a new plant configuration of a biphasic fluid-bed gasification reactor which simplifies the production process of synthesis gas, essentially based on CO and H_2, starting from carbonaceous material.

An object of the present invention, as better described also in the enclosed claims, therefore relates to a process for the production of synthesis gas which comprises:

a. feeding a carbonaceous material having a particle-size lower than or equal to 50 mm, to a gasification reactor;

b. feeding an oxidizing gas to the bottom of the reactor at a flow-rate which is such as to give the solid mass present in the reactor an aspect of a biphasic fluid-bed comprising a dense phase, consisting of the solid and interstitial gas, and a diluted phase consisting of gas bubbles which rise along said bed;

c. filtering the solid particles entrained by the gas, in a cyclone filter positioned internally at the top of the reactor;

d. discharging from the cyclone filter a gaseous stream essentially consisting of synthesis gas and unreacted oxidizing gas; and

e. recycling the filtered solid particles, discharged at the base of the cyclone, directly to the biphasic fluid bed, through an inner duct.

According to the present invention, the biphasic fluid-bed reactor essentially consists of a vertical cylindrical structure suitable for containing the carbonaceous material to be gasified. The vertical structure can be a cylindrical body with a substantially constant transversal section or it can consist of two superimposed cylindrical bodies, with a different transversal section.

The body with the larger transversal section is preferably positioned above the body with the smaller transversal section, whereas a tapered element serves as connection means between the two bodies.

The cylindrical structure with two different bodies is preferred, when the carbonaceous material to be gasified comprises a relatively large particle-size. In this case, in fact, as there is a slowing-down of the gaseous flow rate in the upper part of the reactor, there is consequently a slowing-down of the solid part entrained, with reflux, at least of the larger particle-size, in the fluid bed.

Feeding inlets of the oxidizing gas and

carbonaceous material are envisaged at the bottom of the reactor. The oxidizing gas can consist of oxygen, air, or any other O_2/N_2 mixture in which the oxygen content is higher than 21% by volume, calculated with respect to the dry air. The oxidizing gas can also comprise steam in such quantities as to give an O_2/steam ratio by volume ranging from 0.5 to 3.

The carbonaceous material can consist of coal, or solid oil residues (for example, PET coke), in powder having a particle-size of 1-50 μm, biomasses or mixtures thereof. Preferred carbonaceous material consists of biomass.

The term "biomass", as used in the present description and claims, essentially refers to a product of a biological origin which can be used for energy purposes. Biomasses are therefore all products of agricultural crops and forestation, comprising residues of agricultural processing and silviculture, agro-food waste products destined for human nutrition or zootechnics, residues, not chemically treated, of wood processing and the paper industry, all organic products deriving from the biological activity of animals and human beings, such as those contained in urban waste (waste "organic fraction").

The average chemical composition of a long-stemmed biomass consists of about 25-30% of lignin and 75% of carbohydrates, i.e. molecules of sugar joined to form long polymeric chains. The two most important carbohydrates are cellulose and hemicellulose. The long cellulose polymers form the fibres which give plants their resistance, whereas lignin acts as a glue for keeping these fibres joined. Hemicellulose basically has the role of keeping cellulose and lignin associated.

Other constituents present in biomasses in variable quantities are so-called extracts, compounds with a low molecular weight which can be separated by means of organic solvents (terpenes, fats, waxes, phenols) , or with hot water (tannins and inorganic salts) . The inorganic material, present as a phase closely distributed in the fuel, comprises salts of Si, K , Na, S , CI, P , Ca, Mg and Fe, which produce the ash deposit.

The oxidizing gas is introduced into the gasification reactor in a lower position with respect to the feeding point of the carbonaceous material . In particular, the feeding point of the oxidizing gas is in correspondence with a volume at the bottom of the reactor, called "plenum", from which the oxidizing gas starts its ascent along the height of the reactor. The plenum is separated from the rest of the volume of the reactor by means of a perforated septum on which the material to be gasified at the start-up, rests.

Once the oxidizing gas reaches a sufficient pressure in the plenum to exceed the weight of the overlying mass of carbonaceous material, it begins to flow with a *Ug* rate.

With a *Ug* null, there is no fluid bed: the biomass remains accumulated as in a fixed bed.

By gradually increasing the *Ug,* a progressive increase in the thrust of the gas on the solid is registered, and consequently an increase in the pressure drop ($\Delta\rho$) through the bed. The solid remains stationary under the initial "fixed bed" condition until it reaches the minimum fluidization rate *(Umf)*, in correspondence with which the thrust exerted by the gas on the solid completely balances the weight of the bed.

By increasing the gas rate over *Umf,* the pressure drop through the bed remains substantially constant whereas the bed begins to expand and the slip rate increases together with the vacuum degree. Under these conditions, the system has a biphasic appearance: together with a dense phase consisting of the solid and interstitial gas, there is a diluted phase consisting of gas bubbles which rise along the bed. This is the fluid-dynamic regime in which fluid-bed reactors operate in a boiling configuration.

By further increasing the air introduction rate, the bed configuration gradually changes, reducing its biphasic nature and shifting towards a turbulent state, characterized by a reasonable homogeneity of the distribution of the solid in the gaseous stream: the solid no longer forms a continuous phase, but is dispersed in clusters and in long strips of particles: there is no net upward flow of solid in the bed. The bed remains in turbulent regime until the transport rate *(Utr)*, or blowout rate is reached, close to which a brusque increase in the quantity of particles entrained by the gaseous stream is registered, which would rapidly lead to the emptying of the bed. To maintain the continuity of the process, the entrained particles must be refed from the bottom of the reactor, through their recirculation by means of a specific device. This is the overall fluid-dynamic regime in which circulating fluid-bed reactors operate.

According to the present invention, the preferred fluid-dynamic regime for maintaining the carbonaceous material under stable biphasic fluid-bed conditions can be represented by the following equation, which allows the minimum fluidization rate (*Umf*) to be calculated:

$$\left(\rho_s - \rho_f\right) * g = 150 * \frac{1-\varepsilon}{\varepsilon^3} * \frac{\mu_f}{\left(d'_p\right)^2} * u_{mf} + 1.75 * \frac{\rho_f}{d'_p} * \frac{1}{\varepsilon^3} * u_{mf}^2$$

wherein the densities of the solid matter and of the gas (ρ_s and p_f) respectively, the viscosity of the gas (μ_f), the vacuum degree in the bed (ε) and the average diameter of the solid particles (d'_p) are taken into consideration.

This type of equation is substantially a balance between the force of gravitation and the force which exerts the fluid on the solid. It is in relation to ε, which represents the vacuum degree inside the fluid bed, and is valid for all values of the Reynolds number .

The gas which leaves the bed of carbonaceous material, essentially consisting of synthesis gas and residual non-reacted oxidizing gas, entrains solid particles which are filtered by means of a cyclone arranged internally at the head of the reactor. This arrangement surprisingly not only simplifies the plant configuration but also shows an improved abatement efficiency of the entrained powders.

Various fluid-dynamic tests were carried out for studying the behaviour of a silica bed inside a glass prototype which simulates the geometry of the gasification reactor which is to be used in the process, object of the present invention. A study was effected of the quantity of entrained solid, varying some of the parameters among which:

- the gas flow-rate
- the geometry of the reactor
- the diameter of the particles

so as to be able to effect various evaluations, such as:

- the possibility of seeing the behaviour of the bed;
- the systems to be used for reducing the entrainment of the solid;
- measuring the pressure drops in relation to the type of geometry and in relation to the gas flow-rate.

The prototype has a geometry very similar to that of a gasification reactor.

The quartz column, which simulates the reactor, has an internal diameter of 25 mm and a height of 42 cm, and is equipped with a sintered septum whose function is to uniformly distribute the air and support the material forming the bed.

Air of the supply network is used as gasifying agent, whose pressure and flow-rate are regulated by a pressure reducer and a rotameter (0-2000 1/h). The inlet and outlet pressure are also kept in constant control by means of a differential pressure gauge to determine the pressure drops of the system.

The collection system is obtained by the application of a joint which discharges the entrained solid into a flask. The abatement system of the powders was then obtained with:

- a widening having a diameter equal to 58 mm so as to reduce the gas rate and consequently the entrainment of the solid;
- a cyclone inside the widening ;
- an outer cyclone.

Using various geometries, it was therefore possible to verify the most suitable area, considering two fundamental aspects such as solid entrainment and pressure drops.

The gas flow-rate used in all tests is 640 Nl/h.

Table 1: percentage of entrained product in relation to the geometry used

	A	B	C	D	E
% entrained product	25.8	4.6	3.8	0.3	5.3

wherein:

- A : outer cyclone only
- B : widening + outer cyclone
- C : widening equipped with internal cyclone (outlet of 12 mm) + outer cyclone
- D : widening equipped with internal cyclone (outlet of 3 mm) + outer cyclone
- E : widening equipped with internal cyclone (outlet of 12 mm) with closed side tube.

From the results obtained, it can be observed that the greatest entrainment reduction is obtained with the use of the enlargement. The internal cyclone gives a positive contribution, in particular if the diameter of the cyclone outlet is reduced. In this case the gas does not pass through a small diameter due to the high pressure drops and consequently does not entrain the solid which is blocked by the cyclone.

Pressure drops

During the various tests, the determination of the pressure drops was effected by means of a differential pressure gauge.

The data read (gas flow-rate = 500 1/h) are summarized in Table 2.

Table 2: pressure drops of the various components used

	ΔP read (mbar)
Reactor + filter	63.7
Outer Cyclone + connector	0.02
Widening	0.06
Widening + internal cyclone	0.06

From the data collected, it can be observed that the insertion of the cyclone inside the reactor does not cause a significant increase in the pressure drops already induced by the presence of the widening.

CLAIMS

1. A process for the production of synthesis gas comprising :
   a. feeding a carbonaceous material having a particle size lower than or equal to 50 mm to a gasification reactor;
   b. feeding to the bottom of the reactor an oxidizing gas at a flow rate to be able to provide the solid mass present in the reactor with an aspect of stable biphasic fluid bed, comprising a dense phase consisting of the solid and interstitial gas, and a dilute phase consisting of gas bubbles which ascend along said bed;
   c. filtering the solid particles dragged by the gas, in a cyclone filter placed internally at the top of the reactor;
   d. discharging from the cyclone filter a gaseous stream essentially consisting of synthesis gas and of unreacted oxidizing gas;
   e. recycling the filtered solid particles, discharged at the base of the cyclone, directly in the biphasic fluid bed, by means of an inner duct .
2. The process according to claim 1, wherein the carbonaceous material consists of biomass.
3. The process according to claim 1, wherein the oxidising gas comprises steam (support can be found on page 8 lines 5-7 of the English

Translation) .

4. The process according to claim 3, wherein the 0_2/steam ratio by volume ranges from 0.5 to 3 (support can be found on page 8 lines 5-7 of the English Translation) .

5. The process according to claims from 1 to 4, wherein the stable biphasic bed is represented by the following equation,

$$(\rho_s - \rho_f)*g = 150 * \frac{1-\varepsilon}{\varepsilon^3} * \frac{\mu_f}{(d'_p)^2} * u_{mf} + 1.75 * \text{-^*-!*} \quad u_{mf}^2$$

which allows the velocity of minimum fluidification *(Umf)* to be calculated:
wherein the densities of the solid matter and of the gas, (ρ_s and ρ_f) respectively, the gas viscosity (μ_f), the empty space degree in the bed (ε) and the average diameter of the solid particles (d'_p) are considered.

6. A process according to claims from 1 to 5 wherein the reactor consists of two superimposed cylindrical bodies with a different transversal section (support can be found on page 7 lines 15-16 of the English Translation) .

7. A device for the gasification of a carbonaceous material, comprising a cylindrical vertical reactor, suitable to contain the carbonaceous material to be subjected to gasification, feeding means of the carbonaceous material and oxidizing

gas, placed at the bottom of the vertical reactor, and filtration means of the dragged powders, placed internally, at the head of the reactor.

8. The device according to claim 7 wherein the reactor consists of two superimposed cylindrical bodies with a different transversal section (support can be found on page 8 lines 15-16 of the English Translation) .

9. The device according to claim 8 wherein the body with the larger transversal section is positioned above the body with the smaller transversal section, whereas a tapered element serves as connection means between the two bodies (support can be found on page 8 lines 17-20 of the English Translation) .

10. The device according to claim 7 and 8 wherein the filtration mean of the dragged powders is a cyclone (support can be found on page 7 lines 1-2, page 12 lines 1-4 of the English Translation) .