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Steam-gasification of biomass in a fluidised-bed of olivine particles

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Abstract

Naturally occurring catalytic substances are employed in biomass steam-gasification processes to enhance the yield of fuel gas and reduce its tar content by cracking and reforming the high molecular weight organic components. Calcined dolomite is widely used for this purpose; it exhibits good catalytic activity under the operating conditions of the gasifier. However, due to its poor mechanical strength, it gives rise to a large production of fines in a fluidised-bed environment. This work reports an investigation into the catalytic behaviour of olivine, a common, naturally occurring mineral containing magnesium, iron oxides and silica: iron is known to play a positive role in tar decomposition reactions. The gasification runs, performed with a laboratory scale, biomass gasification unit, show that the olivine activity is close to that exhibited by dolomite under comparable operating conditions. Olivine has the additional advantage, however, that its resistance to attrition in the fluidised bed is much greater, similar to that of sand. Parametric sensitivity studies of a gasification process, utilising olivine as the fluidised-bed inventory, indicate an optimum gasification temperature of just above 800°C, and little influence of the steam/biomass ratio in the range 0.5–1. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Biomass gasification; Tar destruction; Catalytic substances; Fluidisation; Crushed almond shells; Sand, dolomite and olivine bed inventories; Effect of temperature; Steam/biomass ratio

1. Introduction

Since the discovery of fire, biomass has been widely used to obtain heat by direct combustion. It represents a rather low energy-density source, unable to compete with fossil fuels in modern, highly efficient heat and power production systems. It has been found convenient, however, to convert it into a gaseous fuel by means of a high-temperature process known as *gasification*. As a result of rather complex thermo-chemical processes [1], the biomass is thus transformed into permanent gases, such as hydrogen, carbon monoxide, carbon dioxide and methane, together with organic vapours which condense under ambient conditions and are known collectively as *tar*, and a solid residue consisting of char and ash.

Extensive experimental studies reported in the literature [2,3] show that fluidised-bed, steam-gasification processes are capable of maximising the gas product yield as a result of the high heating rates involved, advantageous residence time characteristics, and the efficient tar and char reduction brought about by steam

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reformation. Additional advantages relate to the flexibility of such systems in dealing with a range of solid materials of varying physical property and composition.

The gasification reactions are endothermic overall. In order to provide the necessary heat input, some air is introduced to burn part of the available fuel (biomass, char, and gas). This can give rise to a dilution with nitrogen of the product gas, lowering its calorific value; however, process configurations have been recently developed [4] which attempt to separate the combustion and gasification processes. These involve in principle two parallel fluidised zones, one fed with air, the other with steam; heat transfer from the combustion to the gasification zone is achieved by means of a high circulation rate of the bed inventory through the two zones. At the same time, the design is directed at minimising the contamination of the gasifier product gas with the flue gas from the combustion section.

Such developments in fluidised-bed technology necessitate the adoption of bed inventory materials strong enough to withstand the severe attrition conditions encountered in both the high temperature fluidisation environment and the solid circulation loop, in order to avoid particle fragmentation and a consequential large production of fines with negative consequences for the cyclone and filter processes. The most popular material is sand, which performs very well mechanically, as evidenced by its wide industrial use in bubbling and circulating fluidised-bed combustion applications [5]. Sand, however, plays no active role in the gasification process itself.

In order to render the gaseous product acceptable as a fuel in a wide range of practical applications, the condensable tar must be converted to permanent gases by means of cracking and reforming reactions – which have the additional effect of increasing the gas yield and overall efficiency of the gasification process. This is clearly preferable to the alternative of a scrubbing treatment which is economically unattractive as well as being environmentally problematical. For this reason a number of current research programmes are focused on catalytic processes aimed at significantly lowering the temperature of the tar reforming reactions. Both manufactured catalysts and naturally occurring minerals known to promote tar cracking and reforming have been investigated for potential incorporation in the gasification process [6]. Although the former are undoubtedly needed to reach very stringent specifications on gas purity, there is abundant experimental evidence on the effectiveness of cheap and widely available basic oxide substances in drastically reducing the tar content of gasification products [7]. In fact these are often suggested as preliminary gas conditioning agents, preceding a secondary catalytic reactor in which the gas composition is further *refined* [8].

It is well known that dolomite decomposes tar efficiently at the operating conditions usually employed in gasification processes [9,10], it has been utilised directly in fluidised-bed gasifiers as well as in secondary reactors [11] in both demonstration units and industrial installations [12]. The main problem with dolomite is its fragility. As is well known, natural dolomite undergoes a calcination process at high temperatures [13], so that under gasification conditions (at temperatures typically in the range 800–850°C) the particles are highly porous and fracture easily as a result of attrition phenomena. This gives rise to a large production of fines which, in addition to necessitating a substantial fresh particle make-up, poses problems for the stable operation of the fluidised-bed gasifier and its ancillary units.

Among the factors affecting the activity of the tar decomposition reactions is the positive catalytic role of iron. Studies into the coal gasification processes have long recognised this phenomenon [14,15] and it has been recently confirmed in a study of catalytic tar hydrogenation [16] in which the importance of iron atoms in a tri-metallic perovskite catalyst structure has been demonstrated.

This could well explain the good catalytic behaviour exhibited by olivine, an iron-containing mineral, in a systematic study of bed inventories for the biomass gasification process [17]. It is these findings that led to the present investigation into olivine as a convenient substitute for dolomite in the biomass gasification process. Olivine consists mainly of a silicate mineral in which magnesium and iron cations are embedded in the silicate tetrahedra; it occurs abundantly in many geographic locations. Significant experimental evidence is reported below concerning the catalytic activity of olivine in the destruction of tars, also for its outstanding mechanical strength, comparable to that of sand, even at high temperatures; in this respect its performance is certainly superior to dolomite in a fluidised-bed environment.

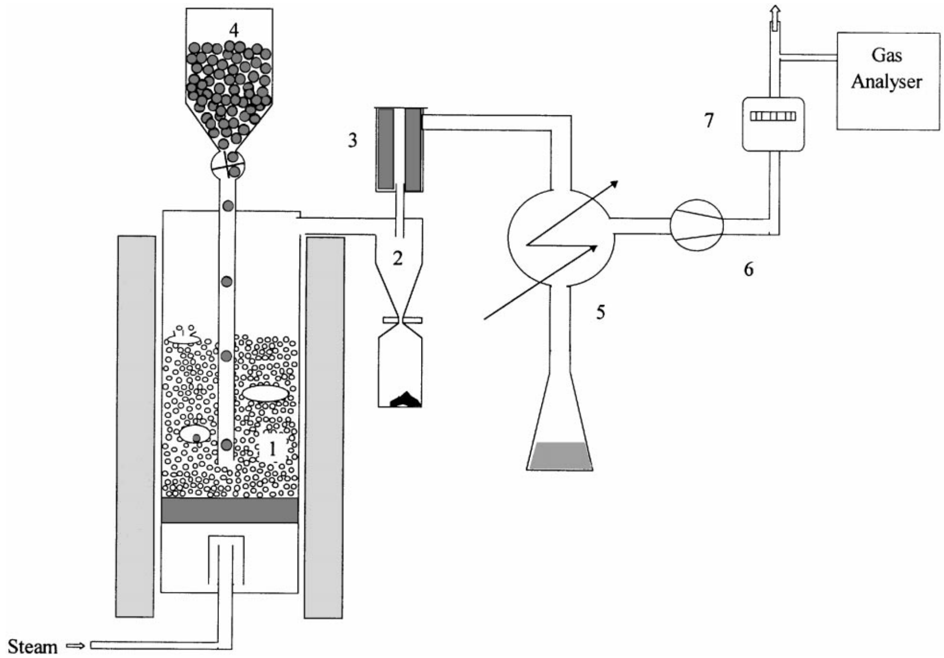

Fig. 1. Schematic description of the experimental rig: (1) fluidised-bed gasifier; (2) cyclone; (3) ceramic candle filter; (4) biomass feeder; (5) water and tar condensation system; (6) blower; (7) volumetric gas-meter.

The experimental tests provide a comparison of the catalytic properties of olivine with those of calcined dolomite in the destruction of tars, when these naturally occurring materials are used as substitutes for sand in a fluidised-bed gasifier. The influence of the main operating parameters on gas yield and composition is also examined.

2. Experimental rig and operating conditions

The experiments were carried out in an experimental rig described in the report of a previous study [16]: only a brief description is given here. It consists essentially of a bubbling fluidised-bed gasifier (60 mm ID) with a continuous biomass feeding system, a gas cleaning section consisting of a cyclone and ceramic candle filter, a cooling system for the separation of water and organic vapours (tar), and various measurement devices. A sketch is provided in Fig. 1.

The fluidising medium is steam, the flow rate of which is controlled by means of a water dosing pump positioned upstream of the steam generator. The bed inventory is either sand, calcined dolomite or olivine particles; their characteristics are reported in Table 1. The olivine batch used in this study was provided by Magnolithe GmbH and consists of a sintered, 'dead burned', crushed and sieved material (MgO 48–50%, SiO_2 39−42%, Fe_2O_3 8–10%), available on the market at about the same price as that for dolomite (120 Eur per metric ton). The amount of sand, dolomite or olivine charged to the gasifier was 600 g for all runs.

The biomass is fed continuously well inside the fluidised bed by means of a jacketed, air-cooled delivery tube; it consists of small round particles of crushed almond shells, chosen because of its relatively high density and amenability to gravity flow in the long, thin delivery tube employed in this small-scale unit. The elemental analysis of the biomass is reported in Table 2, together with average particle size and density. For each gasification run the biomass feed rate was fixed at 0.3 kg h^{-1}; as a result, the weight hourly space velocity (WHSV), expressed as the ratio of the biomass mass flow rate to the weight of the fluidised-bed material, was fixed at 0.5 h^{-1}.

Table 1
Main properties of sand, calcined dolomite and olivine particles (by wt %)

Property	Sand	Calcined dolomite	Olivine
Size distribution (mm)			
> 0.780	0	0	0
0.780–0.655	0	11	6
0.655–0.550	5	33	10
0.550–0.463	20	25	43
0.463–0.390	29	15	20
0.390–0.328	24	11	11
0.328–0.275	14	5	7
0.275–0.231	7	0	3
0.231–0.165	1	0	0
Equivalent diameter (mm)	0.348	0.450	0.410
u_{mf} ($m\,s^{-1}$) (with steam at 820°C)	0.075	0.073	0.09
Particle density (kg/N m^3)	2640	1534	2500

The amount of the organic material condensed together with water is determined by means of total carbon (TC) analysis performed on samples taken every 10–30 min, the instantaneous and average values of the tar content of the gas stream at the exit of the gasifier are calculated assuming naphthalene as the representative tar compound. The overall recovery of water and its conversion are also determined.

The quantity of dry gas produced is measured by means of a volumetric gas-meter. The concentrations of the dry gas are monitored continuously: CO, CO_2 and CH_4 with an infrared analyser, and H_2 with a thermal conductivity detector.

At the relatively low gas velocities adopted ($u/u_{mf} \approx 2.5$) char tends to accumulate mainly in the gasifier; its quantity is therefore obtained by weighing the bed inventory before and after burning off all the carbonaceous residue contained in it at the end of each gasification run.

The experimental tests with sand and dolomite were all carried out at 770°C, with a steam/biomass feed ratio equal to 1; these tests enable a comparison to be made with the catalytic performance of the olivine particles. The experimental runs with olivine were performed at three different values of temperature and of the steam flow rate. In order to reduce the steam/biomass ratio without perturbing the fluidisation quality, sufficient nitrogen was added to the reactor inlet stream to maintain the fluidising velocity at a constant level.

Table 2
Physical and chemical properties of biomass

Type	Almond shells		
Status	Raw	Dry	Dry-ash-free (DAF)
Moisture (wt %)	7.90	—	—
Ash (wt %)	1.16	1.26	—
Volatile matter (wt %)	72.45	78.66	79.67
Carbon (wt %)	46.65	50.65	51.30
Hydrogen (wt %)	5.55	6.03	6.10
Oxygen (wt %)	38.74	42.06	42.60
LHV (kJ kg^{-1})	18350		
Cellulose (wt %)	29		
Hemicellulose (wt %)	28		
Lignin (wt %)	35		
Density (kg m^{-3})	1200		
Particle size (μm)	1100		

3. Results and discussion

3.1. Comparative experimental tests with sand, dolomite and olivine

Table 3 shows the results obtained by performing gasification tests under rigorously identical conditions save for the nature of the bed inventory material. A striking difference in the results is immediately apparent when the inert sand bed is substituted for catalytically active particles: the production of gas increases by more than 50%, with a consequential

Table 3
Comparative experimental tests with sand, dolomite and olivine

Run number	A1	A2	A3
Fluidised-bed inventory	Sand	Dolomite	Olivine
Steam/Biomass	1	1	1
Gasification temperature (°C)	770	770	770
Duration of the gasification test (min)	60	70	60
u/u_{mf}	2.44	2.74	2.75
Average tar content g/N m^3 of dry gas	43	0.6	2.4
Dry gas yield g/N m^3 per kg of DAF biomass	1.1	1.9	1.7
Gas composition (Vol %)			
H_2	43.6	55.5	52.2
CO	33.2	24.0	23.0
CO_2	11.7	14.1	16.9
CH_4	11.5	6.4	7.9
Char yield g per kg of raw biomass	102	30	36
Water conversion (%)	6	39	27
LHV of the gas kJ/N m^3 of dry gas	13 018	10 935	11 369

20 fold reduction in tar content, and over 30% reduction in char. As far as the composition of the permanent gases is concerned, hydrogen increases noticeably (with dolomite its production is more than twice that with sand), whereas methane production maintains more or less the same level. Both of these factors indicate that dolomite and olivine are active for tar destruction but not for methane reforming, as is to be expected. There is also an increase in the production of CO and CO_2 related to the decrease in the char and tar yields.

A check on the mass balance of carbon, hydrogen and oxygen has been performed with reference to each test and the results are shown in Table 4, which includes also the experimental trials examined in the next section of this paper.

In terms of catalytic activity, both dolomite and olivine reveal comparably good performance in terms of the destruction of tar and the consequential increase in the production of permanent gases. This appears more evident when the heating value of the fuel gas produced per kg of biomass is considered (without including the contribution due to the tar components): as reported in Table 3, with olivine it is only 7% less than that obtained using dolomite. It should be stressed that the main purpose for using natural catalytic substances in industrial installations is to effect a substantial reduction in the load of organic vapours in the gaseous product leaving the gasifier, before it comes into contact with a manufactured catalyst which refines the gas purity up to the final requirements; in this respect olivine would appear quite satisfactory, even though its catalytic activity is somewhat lower than that of dolomite.

What is more important is that with olivine the good performance is obtained with a negligible production of fines in the fluidised bed; as we have seen, this represents a major problem with the use of dolomite. The gasification runs performed with sand demonstrated that no noticeable differences exist in the mechanical resistance of sand and olivine particles: the amount of fines collected in the cyclone and filter was found to be negligible in both cases, and the size distribution and average diameter of the particles in the bed inventory did not changed significantly during the course of a test. On the other hand, with dolomite the production of fines gave rise to problems with the smooth operation of the whole system, leading to a steady increase in pressure drop across the filter which became the limiting factor with regard to the duration of a run.

It is worth mentioning at this stage that the conversion level of steam provides a good indication of the extent of the reforming reactions taking place in the gasifier, in particular with regard to the destruction of high molecular weight organic components; this point will be considered further later in next section.

An additional, indirect indication of the catalytic activity of olivine was provided by gasification trials performed by injecting the biomass particles into the free-board of the reactor, instead of using the

Table 4
Mass balance closure for each experiment

Bed inventory (symbol in the figures)	Sand	Dolomite (◇)	Olivine (△)	Olivine (○)	Olivine (■)	Olivine (♦)	Olivine (⊗)	Olivine (⊞)	Olivine (⊕)	Olivine (▲)	Olivine (●)
Gasification temperature °C	770	770	770	700	700	700	820	820	820	820	820
Steam/biomass ratio	1	1	1	0.5	0.7	1	0.5	0.7	1	1	1
Mass Inputs (in g) per kg of raw biomass fed to the gasifier											
C	466.5	466.5	466.5	466.5	466.5	466.5	466.5	466.5	466.5	466.5	466.5
H	175.4	175.4	175.4	119.9	142.1	175.4	119.9	142.1	175.4	175.4	175.4
O	1346.5	1346.5	1346.5	902.0	1079.8	1346.5	902.0	1079.8	1346.5	1346.5	1346.5
Mass outputs (in g) per kg of raw biomass											
Gas C	300.9	382.5	394.1	328.4	293.4	335.3	458.4	452.4	462.8	471.6	494.7
Gas H	59.2	104.9	93.4	59.9	55.3	62.3	87.7	90.1	92.8	96.2	100.2
Gas O	402.7	604.6	624.5	539.5	495.8	591.0	706.6	729.9	785.5	810.0	844.6
Tar C	40.2	1.0	3.4	6.7	4.7	6.8	1.8	1.9	0.9	0.8	1.2
Tar H	2.7	0.1	0.2	0.4	0.3	0.4	0.1	0.1	0.1	0.1	0.1
Char C	102	30	36	150	150.2	105.5	10.0	13	15.6	n.[a]	n.[a]
Water H	112.7	73.1	87.5	57.9	78.1	107.1	34.9	55.0	75.3	74.1	71.9
Water O	901.6	585.1	700.1	463.2	624.6	856.5	279.5	440.4	602.3	592.7	575.5
$\frac{C_{in} - C_{out}}{C_{in}} \times 100$	5.0	11.4	7.1	−4.0	3.9	4.0	−0.8	−0.2	−2.7	−1.3	−6.3
$\frac{H_{in} - H_{out}}{H_{in}} \times 100$	0.5	−1.5	−3.2	1.4	5.9	3.2	−2.3	−1.9	4.1	2.8	1.8
$\frac{O_{in} - O_{out}}{O_{in}} \times 100$	3.1	11.64	1.6	−11.2	−3.8	−7.5	−9.3	−8.4	−3.1	−4.2	−5.5
$\frac{C_{in} + H_{in} + O_{in} - C_{out} - H_{out}}{C_{in} + H_{in} + O_{in}}$	3.3	10.4	2.5	−7.9	−0.8	−3.8	−6.1	−5.6	−2.4	−2.9	−5.0

[a] n. = negligible.

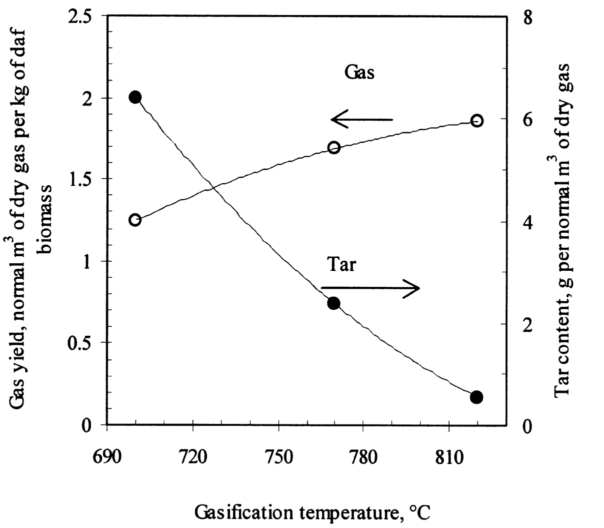

Fig. 2. Gas yield and tar content vs. gasification temperature, with olivine bed inventory.

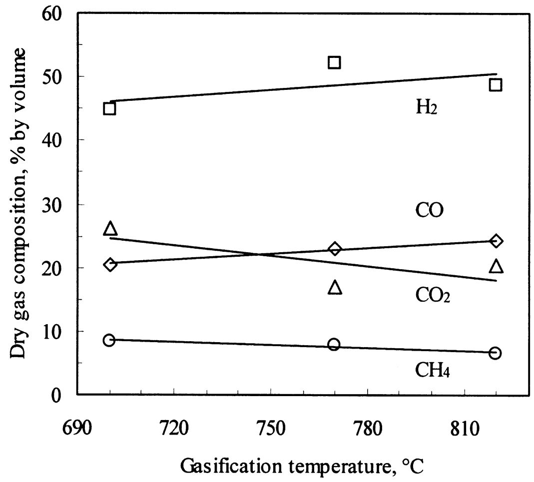

Fig. 3. Dry gas composition vs. gasification temperature, with olivine bed inventory.

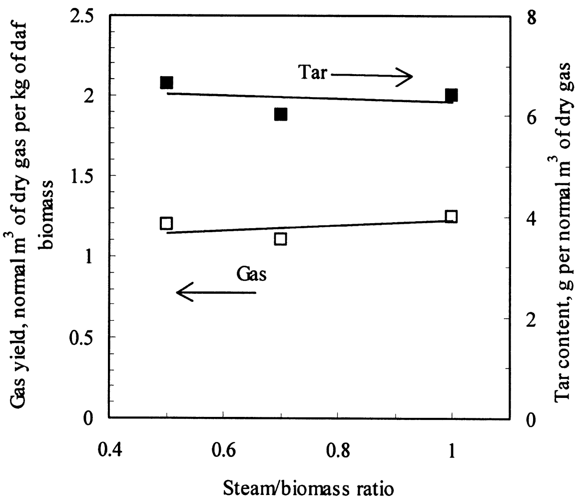

Fig. 4. The influence of the steam/biomass ratio on the production of gas and its tar content; $T = 700^\circ$C.

delivery system described in the previous section. In these tests, which were carried out under otherwise identical conditions, the recorded gas production was found to fall close to that obtained with the standard sand bed, as reported in column A1 of Table 3. It has been stressed in the literature [18] that the relatively low density of biomass (and char) particles precludes good mixing with the bed inventory when the feed point is situated above the bed surface, resulting in a reduced quality of the gaseous product; more negative consequences are of course to be expected when the bed itself is catalytically active.

3.2. *Influence of the operating conditions on the performance of olivine catalyst*

Figs. 2–6 show the production of gas, its tar content, and composition, as functions of the gasification temperature and the steam/biomass ratio for a gasifier bed inventory of olivine particles.

The effect of temperature, summarised in Figs. 2 and 3, has been examined over the range 700–820°C. All the remaining operating conditions were fixed at the values reported previously in this paper. As shown in Fig. 2, the dry gas yield at the maximum temperature was 1.5 times that obtained at 700°C, and the tar content was reduced by a factor of more than 10. The char production, not shown in the figure, was also found to decrease sharply with temperature. These data confirm the good catalytic activity of olivine, and illustrate the importance of operating at gasification temperatures above 800°C. As far as the gas composition is concerned, Fig. 3 indicates that the production of gases with relatively low molecular weights is also favoured by an increase in gasification temperature. The reduction in the methane content is desirable when the end use of the product gas is for feeding fuel cells or as syn-gas for chemical processes.

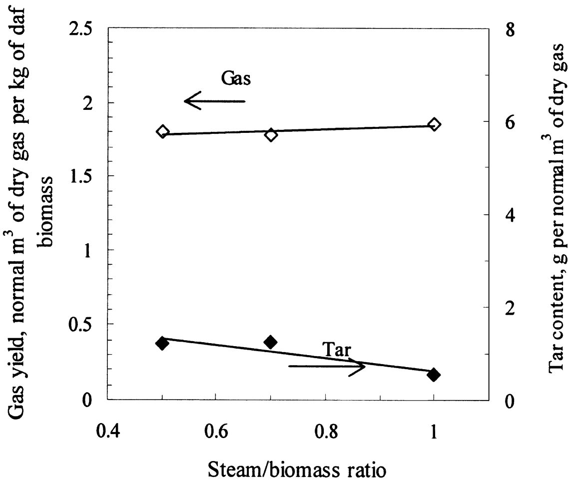

Fig. 5. The influence of the steam/biomass ratio on the production of gas and its tar content; $T = 820°C$.

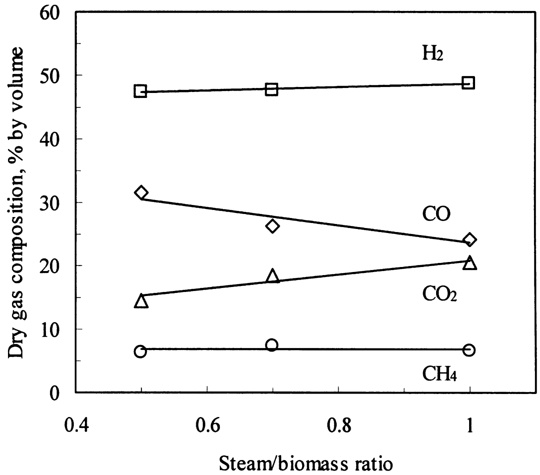

Fig. 6. Dry gas composition as a function of the steam/biomass ratio ($T = 820°C$).

The sensitivity to the steam/biomass ratio has been studied over the range 0.5–1 at the two extremes of the temperature range (700 and 820°C). Figs. 4–6 all show this variable to have a weak effect on gasifier performance, with only a slight increase in gas yield and a slight decrease in tar content with increasing steam flow rate over the interval considered. The gas composition shows the same trend at both temperature levels: the values reported in Fig. 6 were obtained at 820°C. A small reduction in hydrogen concentration, and a corresponding increase in CO concentration, was observed as the steam to biomass ratio was progressively decreased. These results are in good agreement with other published experimental observations [4].

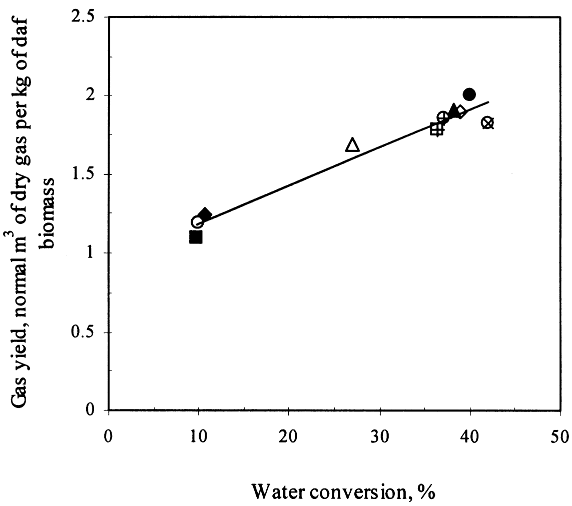

Fig. 7. Gas yield as a function of the steam conversion in the gasifier, at different operating conditions:

Symbol	Bed inventory	Bed temperature (°C)	Steam/ biomass ratio	Particle diameter (mm)
◇	Dolomite	770	1	0.450
△	Olivine	770	1	0.410
○	Olivine	700	0.5	0.410
■	Olivine	700	0.7	0.410
♦	Olivine	700	1	0.410
⊗	Olivine	820	0.5	0.410
⊞	Olivine	820	0.7	0.410
⊕	Olivine	820	1	0.410
▲	Olivine	820	1	0.410
•	Olivine	820	1	0.214

Finally, Figs. 7 and 8 compare the dry gas yields and tar contents, as functions of water conversion level, for gasifier inventories of calcined dolomite and olivine. The correlations revealed in these plots support the suggestion made in the previous section of regarding relative steam consumption as a convenient measure of process efficiency and product quality. It is also worth noting that the experimental runs performed at different steam inlet flow rates (in order to change the biomass/steam ratio), under otherwise similar conditions, gave rise to very similar water conversion values; this suggests that the global rate of reactions involving water is characterised by an approximately linear dependency on steam concentration.

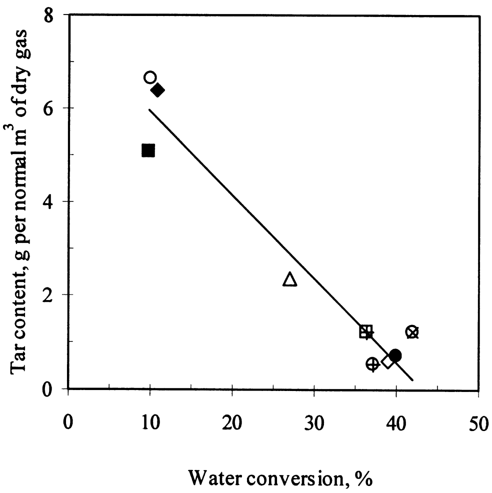

Fig. 8. Tar content in the product gas as a function of the steam conversion in the gasifier; symbols as in Fig. 7.

In Table 5 experimental values of the concentration ratio relevant for the water gas shift reaction are compared with the corresponding theoretical-equilibrium values. Beyond the obvious effect of temperature, these data show that with bed inventories other than sand, the experimental data approach closely the theoretical ones. This is not enough to explain the effect of olivine on the reduction of tar simply in terms of an improved water gas shift reaction: it should be considered that a substantial hydrogen concentration is present in the product gas in each test, including that with a sand bed inventory where such a positive effect on tar destruction is absent. Moreover, it has been observed that the increase of the steam/biomass ratio has only a mild effect on tar conversion obtained at similar operating conditions (Figs. 4 and 5). This last finding is in good agreement with results obtained elsewhere [4] with a quartz sand bed inventory.

As a conclusion we present Fig. 9, where the experimental results of this work are reported together with miscellaneous data published in the literature concerning the use of dolomite to reduce the tar content in the product gas, as functions of temperature and contact ratio, τ. This has been defined as

$$\tau = \frac{\text{kg of calcined dolomite (or olivine)}}{\text{N m}^3\text{exit dry gas/h}}.$$

It should be noted that the value of the contact ratio, τ, in cases where the bed inventory is made of dolomite,

Table 5
Experimental and theoretical-equilibrium values of the concentration ratio relevant for the water gas shift reaction

Bed inventory (Symbol in the figures)	Sand	Dolomite (◇)	Olivine (△)	Olivine (○)	Olivine (■)	Olivine (◆)	Olivine (⊗)	Olivine (⊞)	Olivine (⊕)	Olivine (▲)	Olivine (●)
Gasification temperature (°C)	770	770	770	700	700	700	820	820	820	820	820
Steam/biomass ratio	1	1	1	0.5	0.7	1	0.5	0.7	1	1	1
Moles in the product gas per kg of raw biomass fed to the gasifier											
CO	14.8	18.4	15.8	11.8	9.4	10.4	23.2	19.0	18.3	17.8	19.2
CO_2	5.2	10.8	11.6	11.0	10.8	13.3	10.7	13.3	15.4	16.4	16.8
H_2	19.4	42.6	35.9	20.7	19.1	22.6	34.9	34.2	36.6	37.9	39.6
H_2O	56.3	36.6	43.76	28.9	39.0	53.5	18.7	27.5	37.6	37.0	36.0
$\frac{[CO_2]*[H_2]}{[CO]*[H_2O]}$ (Experimental value)	0.12	0.68	0.60	0.67	0.56	0.54	0.86	0.87	0.82	0.94	0.96
$\frac{[CO_2]*[H_2]}{[CO]*[H_2O]}$ (Theoretical-equilibrium value)	1.14	1.14	1.14	1.52	1.52	1.52	0.95	0.95	0.95	0.95	0.95

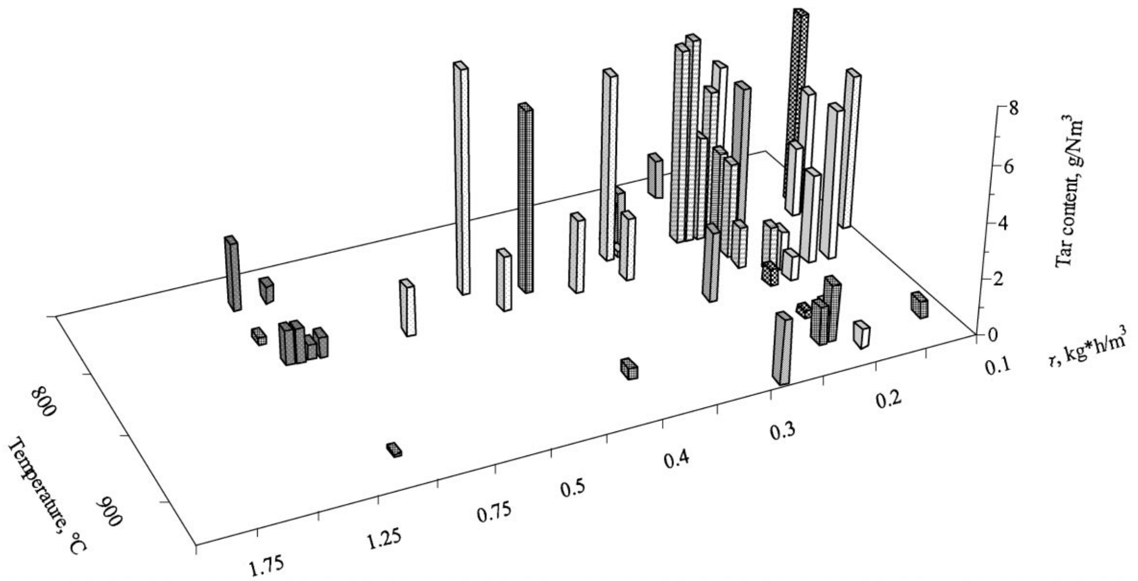

Fig. 9. Tar content in the product gas as function of temperature and contact ratio: Present work ( ) ; Ref. [7] ( ); Ref. [8] ( ); Ref. [15] ( ); Ref. [19] ( ); Ref. [20] ( ), Ref. [21] ( ).

is referred to its calcined product, which is about 60% by weight of the original mass, while olivine does not undergo any weight loss: therefore the differences in τ between this study and most of the investigations with dolomite are effectively smaller than those shown in Fig. 9.

A direct, quantitative comparison among the data in Fig. 9 is out of question, because of differences in operating conditions, gasifying agents, biomass nature and size, process configurations, etc. Nevertheless, such a generalised representation is significative to show the potential of the use of olivine in biomass gasification processes.

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