

High-pressure co-gasification of coal with biomass and petroleum coke

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ABSTRACT

The effects of the main operation variables (temperature, pressure and gasifying agent composition) on gas production and other process parameters, such as carbon conversion, cold gas efficiency and high heating value, during the steam–oxygen gasification of a bituminous coal were studied. It was observed that temperature and oxygen concentration were the most influential variables during the gasification process. In addition, co-gasification tests of binary blends of a bituminous coal with different types of biomass (up to 10%) and petroleum coke (up to 60%), as well as ternary blends of coal–petcoke–biomass (45–45–10%) were conducted in order to study the effect of blending on gas production and carbon conversion.

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1. Introduction

Energy systems based on the use of hydrogen are considered a promising scenario in the long term. Some of the advantages of hydrogen energy include its low environmental impact and its attractive future applications in fuel cells technology for producing electricity [1]. 98% of the hydrogen produced nowadays comes from fossil fuels, mainly from natural gas reforming (approx. 50%) [2]. However, due to the insecurity of its supply and the fluctuations in natural gas prices, it seems that, in the medium term, systems based on coal gasification may offer a possible alternative. Coal gasification is a well established technology for producing syngas ($\text{CO} + \text{H}_2$), where hydrogen production can be increased by means of the water–gas shift reaction ($\text{CO} + \text{H}_2\text{O} = \text{CO}_2 + \text{H}_2$). With this technology, a highly concentrated stream of hydrogen can be generated provided that CO_2 capture is also undertaken. Some studies have shown that electricity generation based on the combination of hydrogen fuel cells and CO_2 capture are less costly compared to post-combustion systems [3,4].

One of the advantages of gasification technology is that energy sources (petcoke, biomass, wastes, etc.), other than coal can be used as feedstock. Petroleum coke is an important by-product of petroleum refineries, and it is employed as feedstock in gasification systems, in combination with coal or separately, to produce a syngas that can be used to satisfy the increasing demand for hydrogen by refineries, and other applications [5]. Moreover, the co-gasification of biomass with coal, could contribute to the reduction of the fossil fuels dependency and CO_2 emissions, as biomass is known to be neutral as regards CO_2

emissions [6]. During co-gasification, coal, biomass and other wastes are put together and converted into a gaseous product stream that can be used to produce electricity, hydrogen, chemicals and liquid transportation fuels. Biomass and wastes are generally more difficult to process than coal, due to their heterogeneity, which produces fluctuations in quality, availability and composition. Therefore, the use of coal with biomass and wastes can provide stable gasification conditions and could prevent problems due to wastes seasonal shortness [7].

On the other hand, biomass has a lower ash and sulphur content than coal, a higher volatile matter yield and fixed carbon with higher reactivity. Therefore, blending low quality coal with biomass and wastes could be attractive from economical, environmental and social points of view in order to make use of possible synergistic effects via the production of fuel gas [8]. Additionally, biomass and coal are considered as potential feedstocks for the supply of syngas (CO and H_2) used in the synthesis of liquid fuels by gasification. In recent years, the advantages of the co-gasification of woody biomass and coal have been reported by several researchers, including the reduction in the production of tar when coal is co-gasified with biomass, which can produce large amounts of tar when gasified alone [6, 9–11].

Currently, co-gasification of coal and biomass is conducted at IGCC electricity generating power stations such as the Willem-Alexander power station in Buggenum (Netherlands), where residual wastes from the agricultural sector, such as sawdust, grape and sunflower seeds, and peanut shells are cogasified with coal. Similarly, at ELCOGAS the world's largest IGCC facility using coal and petcoke as feedstock, located in Puertollano (Spain), there is an ongoing project aimed at evaluating the effects of adding small percentages of biomass, up to a maximum amount of 10%, on the performance of the plant [12].

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Table 1
Proximate and ultimate analyses and high heating values of the samples.

Sample	Proximate analysis (wt.%, db)		Ultimate analysis (wt.%, daf)					HHV (MJ kg ⁻¹)
	Ash	V.M.	C	H	N	S	O*	
PT	37.0	24.3	75.7	5.3	1.4	1.6	16.0	18.7
PC	0.3	9.6	87.6	3.8	1.5	6.2	0.9	35.0
AS	1.2	79.3	49.8	6.1	0.2	0.0	43.9	19.7
OS	0.8	83.8	52.0	6.2	0.1	0.0	41.7	20.3
EB	0.7	83.6	50.6	6.4	0.1	0.0	42.9	19.4

Dry basis (db); dry ash free basis (daf); * calculated by difference.

Within this frame, the effects of co-processing biomass and waste with coal cannot be easily derived from tests at large scale or even at pilot scale, where it is difficult to cover a wide range of conditions, repeat experiments and verify reported trends. Therefore, the results achieved at bench-scale, such as the ones reported in the present paper, are aimed at providing overall trends and to render a framework within which pilot plant operating parameters may be selected.

The objective of this work was to study the effect of several operation variables (temperature, pressure and gasifying agent composition), on gas production, carbon conversion, cold gas efficiency and high heating value, during the steam–oxygen gasification of a bituminous coal. The effect of blending this coal with petcoke and biomass on gas composition was also studied.

2. Experimental

In this work, a bituminous coal (PT), a petcoke (PC), and three types of biomass, almond shells (AS), olive stones (OS) and eucalyptus (EB) were used. The samples were ground and sieved to obtain a fraction with a particle size of 75–150 μm . The proximate and ultimate analyses and the high heating value of the samples are presented in Table 1.

A stainless steel tubular reactor (13 mm internal diameter, 305 mm height) with a porous plate was used for the gasification tests. This reactor is able to work at a maximum pressure of 2 MPa at 1273 K. Fig. 1 shows a flow diagram of the system. Solids are fed continuously into the system from a pressurized hopper. The mass flow rate of the solids is controlled using a pneumatically actuated valve.

The reactor temperature is controlled by means of a thermocouple connected to a temperature controller and data recorder. The thermocouple is in contact with the sample bed. The pressure is measured by a pressure transducer and automatically controlled by a micro-valve. The gasification tests were carried out isothermally at temperatures between 1123 and 1273 K, using a mixture of steam (40–85 vol.%) and oxygen (2–15 vol.%), carried in an inert flow of N₂, at a total flow rate of 200 Ncm³ min⁻¹. The experiments were performed at pressures ranging from 0.5 to 2 MPa. The tars formed during the process and the excess steam were separated from the gas flow by means of a thermoelectric cooler. The gas composition of the dried gases (H₂, O₂, N₂, CO, CH₄ and CO₂) was analysed on-line, using a dual channel micro-GC Varian CP-4900 fitted with a thermal conductivity detector (TCD). The micro-GC

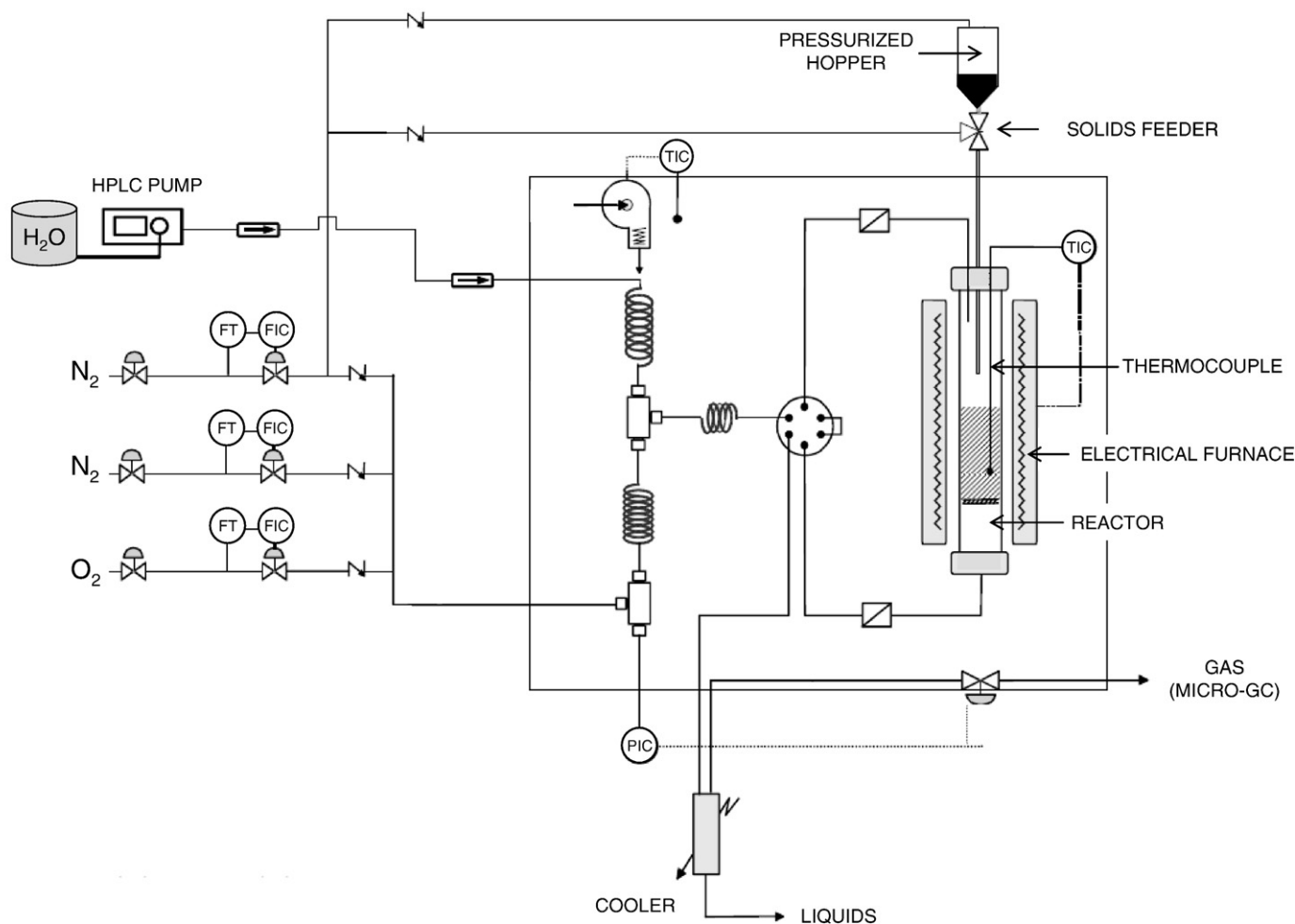


Fig. 1. Flow diagram of the experimental device.

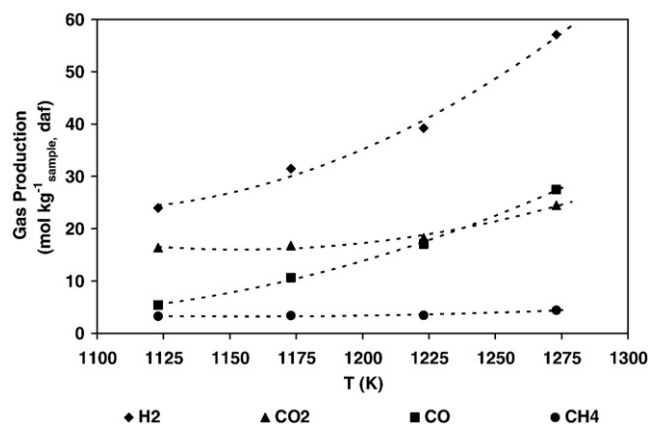


Fig. 2. Effect of temperature on gas production during gasification of coal PT (1 MPa, 5% O₂, 70% H₂O).

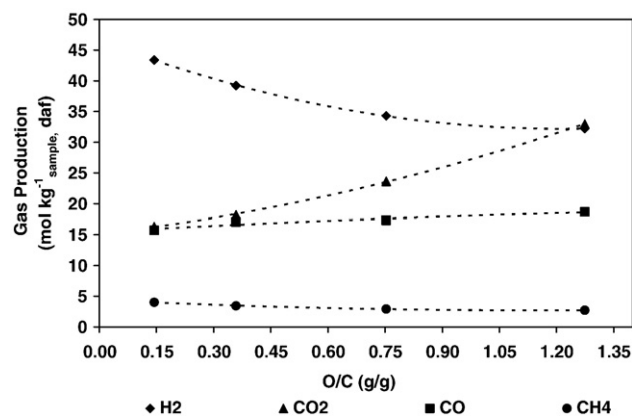


Fig. 3. Effect of the O/C ratio on gas production during gasification of coal PT (1 MPa, 1223 K, 70% H₂O).

was equipped with a molecular sieve, Molsieve 5 Å, and a HayeSep A, columns; helium was used as carrier gas. The system was calibrated employing a standard gas mixture at periodic intervals. The amount of gas generated during the experiments was calculated from a nitrogen balance, since the amount of nitrogen fed in and the composition of the nitrogen evolved are known.

3. Results and discussion

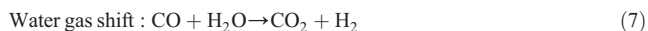
3.1. Effect of operation variables

The final gas composition of the gasification process is the result of the combination of a series of complex and competing reactions:

Oxidation



Water gas



The effect of the main operation variables (temperature, pressure and composition of the gasifying agent) during the steam gasification

of a bituminous coal (PT) on gas production was studied. In addition, other important parameters such as carbon conversion, X (%), gas yield, Y (Nm³ kg⁻¹), gas high heating value, $GHHV$ (kJ Nm⁻³), and cold gas efficiency, η (%), were calculated.

In this work, the carbon conversion, X , was defined as the total carbon contained in the gas produced (CH₄, CO and CO₂) with respect to the total carbon contained in the sample fed in. The gas yield, Y , was calculated as the outgoing dry gas flow rate per mass flow rate of dry fuel. The gas high heating value, $GHHV$, was defined as the gross calorific value of dry gas on a volumetric basis. Finally, the cold gas efficiency, η , was defined as the ratio between the energy content of the gas and the energy contained in the solid fuel.

3.1.1. Reaction temperature

Reaction temperature is one of the most important operating parameters affecting the performance of gasification, due to the balance between endothermic and exothermic reactions involved [7,13]. To study the effect of temperature, the reactive gas composition was set at 5% O₂ and 70% H₂O, according to a previous optimization procedure aimed at maximising the production of H₂ + CO and increasing the carbon conversion [14,15]. The pressure of the system was fixed at 1 MPa. Fig. 2 shows the influence of the bed temperature on gas production. An increase in the temperature clearly favours the formation of H₂ and CO, since the endothermic gasification reactions (Eqs. (3)–(5)) are enhanced. However, at low temperatures, carbon oxidation reactions (Eqs. (1) and (2)) are more significant than carbon gasification, since the former are exothermic, and CO₂ production is more favoured. As the reaction temperature increases, carbon tends to react with CO₂, through the Boudouard reaction (Eq. (3)), displacing the equilibrium to CO formation [16], as can be seen in the evolution of the CO/CO₂ ratio (Table 2). The increase in CO affects the H₂/CO ratio which falls from 4.4 to 2.1 as the temperature increases from 1123 to 1273 K. The production of CH₄ remains almost constant and is not affected by the variation in reaction temperature. This indicates that the methanation reaction (Eq. (6)), does not take place to any

Table 2
Gasification parameters of coal PT at different temperatures (1 MPa, 5% O₂, and 70% H₂O).

	T (K)			
	1123	1173	1223	1273
H ₂ /CO	4.4	3.0	2.3	2.1
CO/CO ₂	0.3	0.6	0.9	1.1
GHHV (kJ Nm ⁻³)	5402	6517	7059	7586
Y (Nm ³ kg ⁻¹)	1.3	1.5	1.7	2.3
η (%)	38.0	50.6	64.3	94.4
X (%)	39.7	48.9	61.4	89.4

Table 3
Gasification parameters of coal PT at different O/C and H₂O/C ratios (1 MPa, 1223 K).

	O/C				H ₂ O/C			
	0.14	0.36	0.75	1.27	1.47	2.07	2.86	4.03
H ₂ /CO	2.8	2.3	2.0	1.7	2.0	2.2	2.3	2.3
CO/CO ₂	1.0	0.9	0.7	0.6	1.2	1.0	0.9	0.8
GHHV (kJ Nm ⁻³)	7182	7059	6793	6448	4854	5987	7059	8588
Y (Nm ³ kg ⁻¹)	1.8	1.7	1.6	1.7	2.1	1.9	1.7	1.4
η (%)	68.8	64.3	58.3	57.0	55.7	61.8	64.3	63.8
X (%)	57.2	61.4	69.6	86.2	52.7	58.2	61.4	63.2

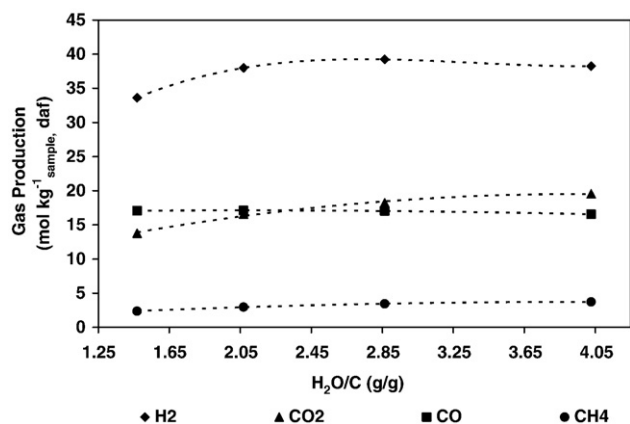


Fig. 4. Effect of the H_2O/C ratio on gas production during gasification of coal PT (1 MPa, 1223 K, 5% O_2).

significant extent and CH_4 is produced mainly during the pyrolysis step [14].

A rise in the reaction temperature produces an increase in carbon conversion through oxidation and gasification reactions (Eqs. (1)–(6)). This originates an increase in gas yield and its HHV, which leads to a higher cold gas efficiency. These results agree with the studies published by other authors who employed different types of gasifiers such as pressurized spouted bed and fluidised bed reactors [13,17–20]. The carbon conversion undergoes a considerable increase (28%) between 1223 and 1273 K; cold gas efficiency also increases reaching a value of 94.4% at 1273 K (see Table 2).

To evaluate the differences between the experimentally obtained gas concentrations and those corresponding to the equilibrium, a gasification reaction equilibrium model based on the minimization of the total Gibbs free energy was used [21]. It was observed that for all the temperatures tested in the present work, the experimental H_2 and CO concentrations were lower than the predicted values, while CO_2 concentrations were higher than the theoretical results. However, the differences between the experimental and equilibrium values diminished as the temperature increased. For instance at 1123 K the experimental values for the concentrations of H_2 and CO were 25.7% and 5.8%, respectively, while those of the equilibrium were 43.8% and 18.3%, respectively. However, at 1273 K the experimental values were 34.6% (H_2) and 16.6% (CO), while the equilibrium values were 42% (H_2) and 18.8% (CO). Among the reasons that can cause the difference between the equilibrium and the experimental results is the carbon conversion, as it has been stated by other researchers [7,22,23]. Higher temperatures improve the extent of gasification reactions, and

Table 4

Gasification parameters of coal PT at different pressures (1223 K, 5% O_2 , and 55% H_2O).

	P (MPa)			
	0.5	1.0	1.5	2.0
H_2/CO	2.2	2.2	2.3	2.3
CO/CO_2	1.2	1.0	1.0	1.0
HHV ($kJ\ Nm^{-3}$)	6384	5987	6223	6073
$Y\ (Nm^3\ kg^{-1})$	2.0	1.9	2.1	2.1
$\eta\ (\%)$	67.5	61.8	69.0	67.3
$X\ (\%)$	60.9	58.2	62.7	61.4

therefore, result in gas composition closer to equilibrium. In this way a lower difference between experimental concentrations and theoretical ones is attained.

3.1.2. Gasifying agent composition

The composition of the gasifying agents determines that of the gas produced and, in turn, the efficiency of the process. In this work, this parameter was studied by varying the oxygen and steam concentrations in the reactive gas, and expressing the results as a function of the O/C and H_2O/C ratios. Fig. 3 shows the production of the main gases during the gasification of coal PT (1223 K and 1 MPa) using a steam concentration of 70 vol.% ($H_2O/C = 2.86$), as a function of the O/C ratio. As a result of the increase in oxygen content in the gasifying agent, CO_2 production increases sharply, mainly due to coal oxidation. The significant decrease in H_2 production is a consequence of oxygen being much more reactive to carbon than steam [24]. What is more, some of the H_2 produced might have burned up if oxygen were present. For this reason, although an increase in the O/C ratio produces an increase in the conversion of carbon, there is a concomitant decrease in cold gas efficiency due to the lower HHV of the gas produced (see Table 3) [7,25]. In the results shown in Fig. 3 there is a slight reduction in CO production, while CH_4 remains practically unaltered, in accordance with the results obtained by other authors employing other types of reactor [7,13].

The effect of varying the steam concentration (H_2O/C ratio), employing an oxygen concentration of 5 vol.% ($O/C = 0.36$), on gas production is shown in Fig. 4 (1223 K, 1 MPa). Carbon steam gasification (Eq. (5)) is enhanced when the H_2O/C ratio increases, leading to a slight increase in H_2 and CO_2 production, while CO production remains almost constant. This suggests that the secondary steam gasification reaction is favoured to the detriment of the primary one at high steam concentrations. At high values of H_2O/C ratio, H_2 production decreases slightly, which could be due to the formation of CH_4 (Eq. (6)). The theoretical gas composition, calculated from the equilibrium model, shows a trend similar to the experimental values.

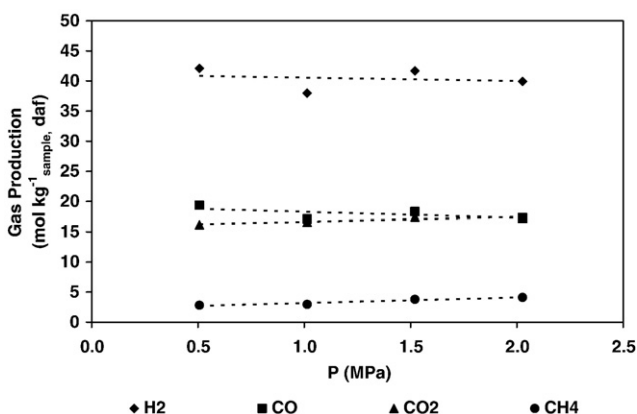


Fig. 5. Effect of pressure on gas production during gasification of coal PT (1223 K, 5% O_2 , 55% H_2O).

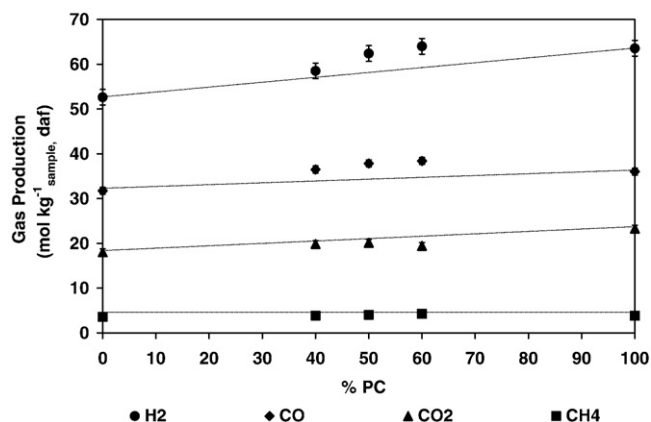


Fig. 6. Gas production during gasification of PT-PC binary blends (1.5 MPa, 1273 K, 5% O_2 , 55% H_2O).

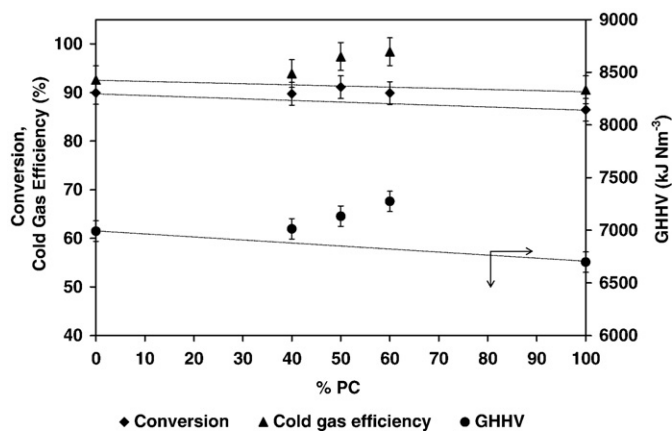


Fig. 7. Carbon conversion, cold gas efficiency and high heating value obtained during co-gasification of PT–PC binary blends (1.5 MPa, 1273 K, 5% O₂, 55% H₂O).

This result confirms that the increase in steam in the gasifying medium favours the secondary steam gasification reaction (Eq. (5)) to the detriment of the primary reaction (Eq. (4)).

As can be seen in Table 3, the H₂/CO ratio increases slightly as the steam content rises in the gasifying agent. However, this parameter clearly decreases when there is a rise in the concentration of oxygen in the reactive gas. What is more, carbon conversion is affected more by the variation in O/C ratio than by the H₂O/C ratio, as can be observed in Table 3. These findings could be useful when deciding the composition of the gasifying agent depending on the application

Table 5

Gasification parameters of binary blends of coal PT with biomass (1.5 MPa, 1223 K, 5% O₂, and 55% H₂O).

% Biomass	AS			EB		OS	
	0	5	10	5	10	5	10
H ₂ /CO	2.3	2.0	2.1	2.0	2.2	2.0	2.2
CO/CO ₂	1.0	1.2	1.2	1.1	1.0	1.2	1.1
GHHV (kJ Nm ⁻³)	6223	6363	6276	6234	5882	6579	6575
Y (Nm ³ kg ⁻¹)	2.1	2.2	2.3	2.4	2.6	2.3	2.3
η (%)	69.0	75.2	77.7	78.2	80.2	80.2	78.9
X (%)	62.7	69.2	71.0	73.9	74.4	72.6	70.1

desired of the gas produced (electricity, hydrogen production, liquid fuels synthesis through Fischer–Tropsch method, etc.).

3.1.3. Reaction pressure

The effect of reaction pressure on gas production during the gasification of coal PT was studied and the results are presented in Fig. 5 (1223 K, 5% O₂, 55% H₂O). In this figure it can be observed that, in the range of pressures studied in this work, pressure does not have a significant effect on gas production. However, an increase in pressure was observed to produce a slight increase in CH₄ and CO₂ to the detriment of H₂ and CO [19], as the increase in pressure shifts the equilibrium to the side with the fewer moles of gas (Eqs. (8) and (9)). The same tendencies were observed when the equilibrium gas composition was calculated by varying the total pressure. It can be seen in Table 4 that only the high heating value of the raw gas is reduced slightly due to the change in pressure.

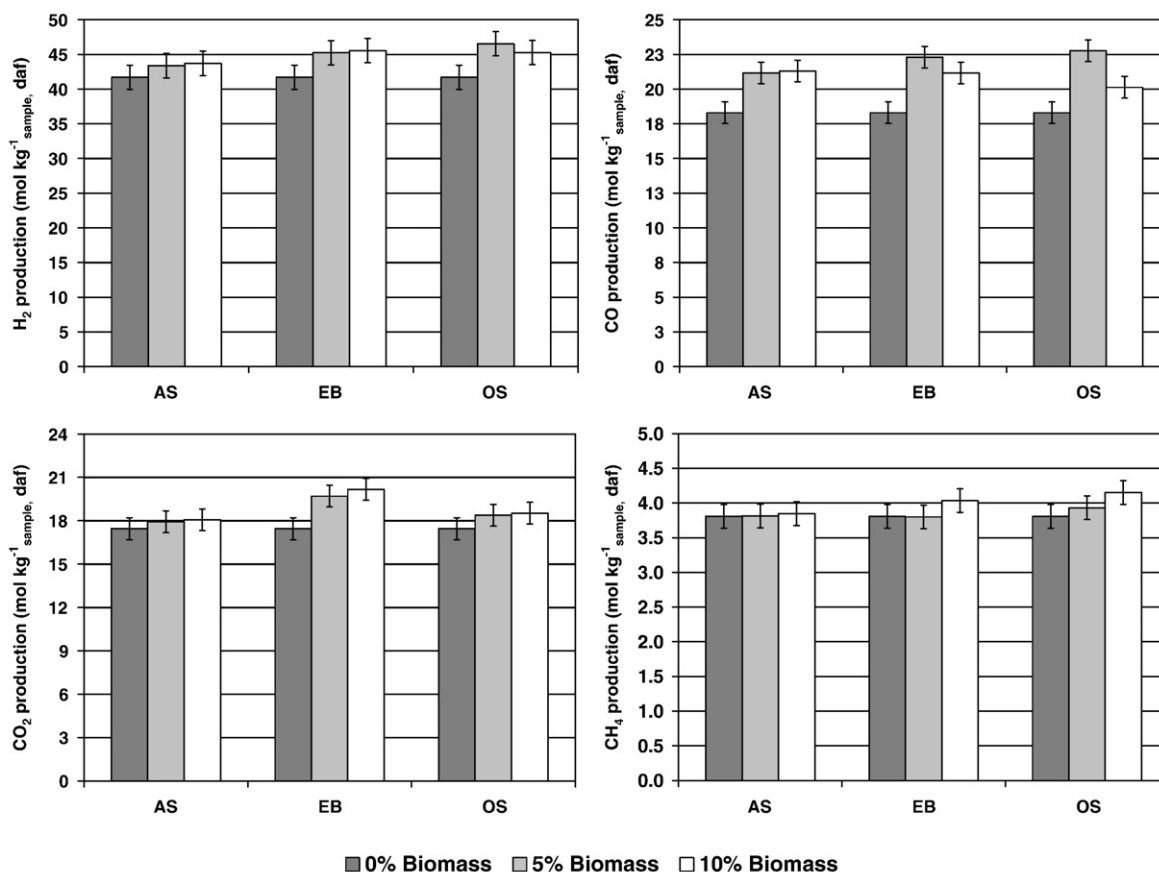


Fig. 8. Gas production during co-gasification of binary blends of coal PT with biomass (1.5 MPa, 1223 K, 5% O₂, 55% H₂O).

3.2. Effect of blending fuels

The effect of blending coal with biomass and petcoke was studied in order to determine the possible presence of synergetic phenomena. Gasification tests of binary blends of coal PT combined with different biomass samples (AS, EB and OS), and with petcoke (PC), as well as ternary blends (PT–PC–Biomass) were carried out. For this purpose, the feed mass flow rate was set to maintain the values of the O/C and H₂O/C ratios constant. The experimental conditions were fixed at 5% O₂ and 55% steam, with a pressure of 1.5 MPa and a temperature of 1273 K. The experimental error margin was evaluated by calculating the errors produced in the gas composition from repeating an experiment several times on different days. The values obtained, expressed in mol of gas per kg of sample on a dry ash free basis, were lower than 2 for H₂, and lower than 1 for CO and CO₂.

Fig. 6 shows the main gases production during the gasification of the PT–PC blends. There is a positive deviation from the linear additive rule in the case of H₂ and CO production, whilst the amount of CO₂ and CH₄ can be calculated from the production of the individual blend components and proportions. These results show a positive synergetic effect between the components of the blend. This type of behaviour can also be observed in the results shown in Fig. 7, where carbon conversion, cold gas efficiency and high heating values are plotted as functions of the percentage of the PC in the blends. The carbon conversion decreases with the increase in petcoke content due to its lower reactivity compared to coal [26]. However, in all cases a clearly positive effect was attained when coal PT was blended with petcoke PC.

In the case of the blends of PT with different biomasses (Fig. 8) the production of H₂, and especially that of CO, increased with additions of up to 10% biomass. The increase in CO₂ was more evident when coal was blended with EB. This behaviour in gas production was reflected in the H₂/CO ratio, which decreased slightly for all the blends (see Table 5), in agreement with the results found by other authors [11,27]. In general, other process parameters such as carbon conversion and cold gas efficiency clearly improved when coal PT was gasified with biomass due to the high carbon conversion to gas.

Finally, ternary blends of PT, PC and the three biomasses were gasified to determine the presence of possible interactions between the three fuels due to the positive effects observed when PT–Biomass and PT–PC were blended separately. Fig. 9 shows the gas production during the gasification experiments of the PT–PC–Biomass ternary blends (45–45–10%). In this figure it can be seen that when biomass was used with blend PT–PC, H₂ production remained almost constant. However, a slight decrease in CO production and a slight increase in CO₂ production were attained in all cases. This is reflected in the

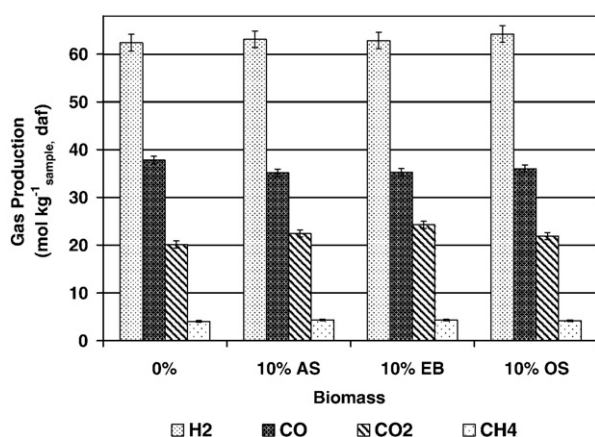


Fig. 9. Gas production during co-gasification of ternary blends of PT–PC (45–45%) with biomass (10%) (1.5 MPa, 1273 K, 5% O₂, 55% H₂O).

Table 6

Gasification parameters of ternary blends of PT–PC (45–45%) with biomass (10%) (1.5 MPa, 1273 K, 5% O₂, and 55% H₂O).

% Biomass	AS	EB	OS
	10	10	10
H ₂ /CO	1.6	1.8	1.8
CO/CO ₂	1.9	1.6	1.6
GHHV (kJ Nm ⁻³)	7132	6779	6607
Y (Nm ³ kg ⁻¹)	3.7	3.9	4.0
η (%)	97.4	101.4	101.3
X (%)	91.1	94.8	97.5

evolution of the H₂/CO and CO/CO₂ ratios presented in Table 6. This table shows an improvement in the efficiency of the process due to the increase in carbon conversion and cold gas efficiency.

4. Conclusions

The effect of temperature, pressure and gasifying agent concentration on gas production and other process parameters such as carbon conversion, cold gas efficiency and high heating value was studied. It was found that temperature and oxygen concentration had the greatest influence on the final products, which were practically unaffected by changes in reaction pressure. An increase in the temperature of reaction led to a significant increase in H₂ and CO production. With respect to the O/C ratio, it was observed that the production of CO₂ and CO increased to the detriment of H₂, which decreased sharply. When the concentration of steam in the reactive gas was increased, an increase in H₂ production was observed, whereas CO production diminished slightly. A synergistic effect was observed for blends of coal with petcoke, and an increase in the production of H₂ and CO was obtained. Furthermore, the addition of a small amount of biomass (up to 10%), led to an increase in H₂ and CO production in all the biomasses used. Finally, blending biomass with PT–PC blends did not produce any significant change in H₂ production, although slight variations were observed in the production of CO and CO₂.

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