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Catalytic Co-gasification of Lignites Blended with a Forest Residue under the Carbon Dioxide Stream

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Abstract

The present study investigated the co-gasification of two different lignites blended with the forest residue collected from the land restoration activity sites of open-pit mines located in the region of the Ptolemais basin in North Greece performed under the carbon dioxide stream. All samples were devolatilized in a fixed bed unit prior to the gasification evaluations. The gasification evaluations were performed using a thermal analysis system (TG/DTG) operated at temperatures of up to 1000 °C. The reactivity, conversion, cold gas efficiency, and influence of the external catalysts CaO and K_2CO_3 were assessed in the evaluations. The reaction rate of the forest residue was 2–3 folds higher than that of the lignites, with the conversion of the former reaching a value of 96.4% (dry basis), while the conversion of the lignites varied between 43.4% and 51.6%. The peak inflection temperature was in the range of 859–939 °C. The reactivity of the lignite/biomass blends was higher than that of the lignites, and the final conversion was increased by approximately 30%. When individual biochars were impregnated with 30% CaO or K_2CO_3, the process occurred at lower temperatures, and the conversion of the lignites increased by 35%–40% while that of the forest residue reached a value of 100%. The CaO catalyst performed better. Finally, a blend of equal amounts of Kardia lignite or Ahlada lignite and the forest residue with 30% CaO was formulated, which resulted in an 89.6% or 71.7% conversion to carbon monoxide gas, respectively.

Keywords
Co-gasification; lignite; forest residue; catalysts

1. Introduction

Coal is the fossil fuel with a major feedstock for electricity generation worldwide, accounting for approximately 40% of the global power production [1-3]. In Greece, mineable brown coal deposits amount to 4.6 billion tons, of which approximately 50 million tons are burned in mine-mouth power plants every year. However, recent years have witnessed a depletion in the coal reserves and massive emissions of greenhouse gases contributing to global warming, which have together affected the power generation sector greatly, resulting in increased operating costs of coal units and, consequently, lower consumption [4]. Therefore, to maintain the economic sustainability of power plants, their operating flexibility must be increased, which could be achieved by co-processing coal with renewable sources of energy. In this context, biomass materials, which comprise a wide range of species, appear to be an attractive choice for the partial substitution of coal owing to their abundance, carbon neutrality, and low-cost availability [5, 6].

A combination of coal and biomass, when used as feedstock for power production, offers environmental benefits, in terms of reduction in the amounts of produced emissions and biodegradable wastes dumped in landfills, as well as economic benefits, including a reduced requirement for large investments in stand-alone biomass plants that would have otherwise suffered the lower energy density of biomass and its seasonable availability [2, 7, 8]. In addition, the combined use of coal and biomass enables fulfilling the global policies established for de-carbonized energy and circular economy [9]. In the process of fuel combustion, the gasification process offers several advantages over direct combustion, such as higher efficiency, lower emissions, and convenient control and flexibility of the feedstocks and end products [6, 10-12]. Air, when used as the gasifying agent, produces a lower heating value of the product gas, while a mixture of H_2, CO, and CO_2 gases is generated in the presence of steam. The produced syngas may then be utilized in internal combustion engines, gas turbines, solid oxide fuel cells, and the synthesis of several biofuels and chemicals [1, 13]. The implementation of the post-treatment technique of CO_2-char gasification, in which the residual CO_2 streams are converted to marketable fuels rather than being captured and sequestrated, has long-term benefits of mitigating greenhouse gas emissions and the concomitant production of clean fuels [10, 11]. In addition, pyrolysis when performed as a pre-step to gasification produces a highly reactive char that is free of tar and the potential consequent operational problems, along with bio-oil and pyrolytic gas, which serve as valuable sources of energy [6, 11].

The gasification of chars using carbon dioxide at a low heating rate is a slow process, so that their reactivity must be increased to ensure enhanced conversion. Several factors affect char reactivity in the gasification process, among which the reaction temperature, the structural and chemical characteristics of fuels used, and the composition of ash are the most important ones. A higher pyrolysis temperature, feedstock aromaticity, and microporosity reportedly lower the reactivity of chars [10, 11, 14, 15]. On the contrary, a higher number of active sites on the char surface and the presence of alkali or alkaline earth metals in the forms of carbonates or hydroxides are reported to enhance the reaction rate [10, 14, 16, 17]. In the co-gasification process, the alkali minerals inherent in the biomass serve as natural catalysts, thereby reducing input costs [1, 11, 15, 18, 19].

The co-gasification of high-ranking coals with biomass has been investigated intensively [1, 11, 15, 18, 19]. Most of these studies have addressed the synergetic effects, which were attributed to the alkali metals present in the biomass ash [1, 2, 18, 19]. However, no consensus exists, to date, on the most suitable factor for the evaluation of the gasification reactivities of different carbonaceous materials. The use of different coal types or biomass materials could, therefore, lead to different kinds of interactions during co-processing. In regard to the CO_2–char co-gasification of low-ranking coals, such as lignite with forest residues, and the use of external catalysts for promoting reactivity, little information exists in the current literature, to the best of the author's knowledge.

In this context, the present study evaluated the co-gasification performance of two different lignites blended with a forest residue obtained from the land restoration activity sites of different mines under a carbon dioxide stream to determine the reactivity, conversion, cold gas efficiency, and influence of external catalysts.

2. Materials and Methods

2.1 Raw Materials, Preparation, and Characterization

The samples used in the present study were provided by the Public Power Corporation of Western Macedonia, Greece. Two lignites obtained from the open-pit mines of Kardia (LK) and Ahlada (LA), located in the region of the Ptolemais basin, and Acacia waste wood (AW) obtained from the land restoration activity sites of the mines were used. The lignites were selected according to their quality and their calcite content. It was determined subsequently in the study, as would be described below, that the Kardia lignite was rich in calcite, as opposed to the Ahlada lignite, and also had a higher calorific value. Raw samples were air-dried, riffled, ground, and sieved to a particle size below 250 μm. A jaw crusher and a ball mill were used for grinding the lignites, while a cutting mill was used for grinding the biomass material. Representative samples were characterized by performing the proximate analysis, ultimate analysis, and calorific value determination. The characterizations were based on the ASTM standards for lignites (D5142, D5373, D4239, and D5865) and the European standards (CEN/TC335) for acacia waste wood. Mixtures of each lignite with the biomass material in a 50:50 (wt./wt.) ratio were prepared and characterized using the standard methods stated above.

A chemical analysis of the produced ashes was performed using an X-ray fluorescence spectrometer (model S2 Ranger EDS, Bruker AXS).

Prior to the gasification experiments, the samples were devolatilized in a stainless steel fixed bed unit (ID = 70 mm, H = 140 mm), described in detail in a previous report [6]. The flow of nitrogen was 150 mL/min, the heating rate was 10 °C/min, the final temperature was 600 °C, and the retention time was 30 min. The produced biochars were characterized as described earlier.

The calcium oxide and potassium carbonate, which were purchased from Sigma-Aldrich, were used as the gasification catalysts in the incipient wetness impregnation method. The concentration range used was 10%–30% (wt. %), which corresponded to 0.1–0.5 gMe/gC in char [6, 17, 20].

2.2 Gasification Experiments

The gasification experiments were conducted using a thermogravimetric/differential thermogravimetric (TG/DTG) analyzer to monitor weight loss and the rate of weight loss as functions of temperature and time under dynamic conditions. The gasification evaluations were performed at a precision temperature of ±2 °C and a microbalance sensitivity of <5 µg. Each biochar was gasified under a carbon dioxide atmosphere with a flow rate of 35 mL/min and a heating rate of 10 °C/min, up to 1000 °C, until a constant weight was achieved. Nitrogen was used as the purge gas at a flow rate of 45 mL/min. The experiments were conducted under chemical control (sample mass ~ 15 mg and particle size < 250 µm) and repeated two or three times to evaluate the reproducibility of the results, which was expressed in terms of the relative standard deviation (RSD).

The initial T_i, the peak inflection temperature T_{max}, the reaction rate R, and its peak R_{max} were derived from the thermograms, while the reactivity was calculated from the following equation:

$$R_f = \frac{R_{max}}{T_{max}} \times 100 \ (min^{-1}/C) \tag{1}$$

The conversion of the process was expressed on a dry basis and was calculated using the equation provided below:

$$\alpha = \frac{m_o - m}{m_o - m_f} \times 100 \ (\%) \tag{2}$$

In the above equation, m_o, m, and m_f denote the initial, current, and final masses of the biochar.

The cold gas efficiency was calculated according to the following equation:

$$CGE = \frac{m_g LHV_g}{m_b LHV_b} \times 100 \ (\%) \tag{3}$$

where, m_g and m_b denote the masses of the gas and biochar, respectively (kg), while LHV_g and LHV_b denote the lower heating values of the gas and biochar, respectively (MJ/kg).

3. Results and Discussion

3.1 Characterization of the Raw Fuels and Biochars

The results for the proximate analysis and ultimate analysis of raw fuels and biochars are presented in Table 1. As visible in Table 1, acacia waste wood contained a much higher concentration of volatile matter (85%) compared to lignites and a much lower ash content (2.1%). Accordingly, the higher carbon and hydrogen contents of acacia waste wood, together with the lower ash content, resulted in a higher calorific value. The sulfur and nitrogen contents were low in all samples, which implied reduced toxic emissions during the gasification process.

Table 1 Results of the proximate analysis and ultimate analysis of raw fuels and biochars (% dry).

Sample	Volatile matter	Fixed carbon	Ash	C	H	N	O	S	HHV* (MJ/kg)
LK_r/LK_b	43.6	23.1	33.3	36.0/41.4	2.9/1.2	1.2/1.1	25.9/4.7	0.7/0.2	18.0/30.2
LA_r/LA_b	36.4	27.5	36.1	27.3/38.6	2.1/1.2	0.6/0.6	32.8/5.5	1.1/1.3	10.9/29.0
AW_r/AW_b	85.0	12.9	2.1	49.0/79.9	6.6/1.7	1.0/1.0	41.3/8.3	-	19.7/30.6

* dry-ash-free; r = raw; b = biochar.

Table 1 also reveals that after devolatilization at 600 °C, all materials were enriched in carbon, while their hydrogen and oxygen concentrations were decreased due to the removal of H- and O-bearing compounds upon the thermal treatment. Accordingly, the higher carbon content on the one hand and the lower oxygen content on the other raised the calorific value of the biochar samples to approximately 2–3 fold the original value of raw fuels. In addition, no N- or S-containing volatile species were evident up to 600 °C.

The chemical analysis of the samples was performed to reveal their major inorganic elements, which were expressed in the form of oxides. The results are illustrated in Figure 1. It was observed that the Kardia lignite and Acacia waste wood were rich in Ca and Si, while the Ahlada lignite contained a high concentration of Si and a low of Ca. The two lignites contained considerable amounts of Al and Fe as well. The concentrations of K and Na were quite low in all fuels. According to previous studies [6, 12, 16, 17, 21], the alkali metals K, Na, Ca, Mg, and Fe exhibit catalytic activity during the gasification of char, while Si and Al decelerate the reaction.

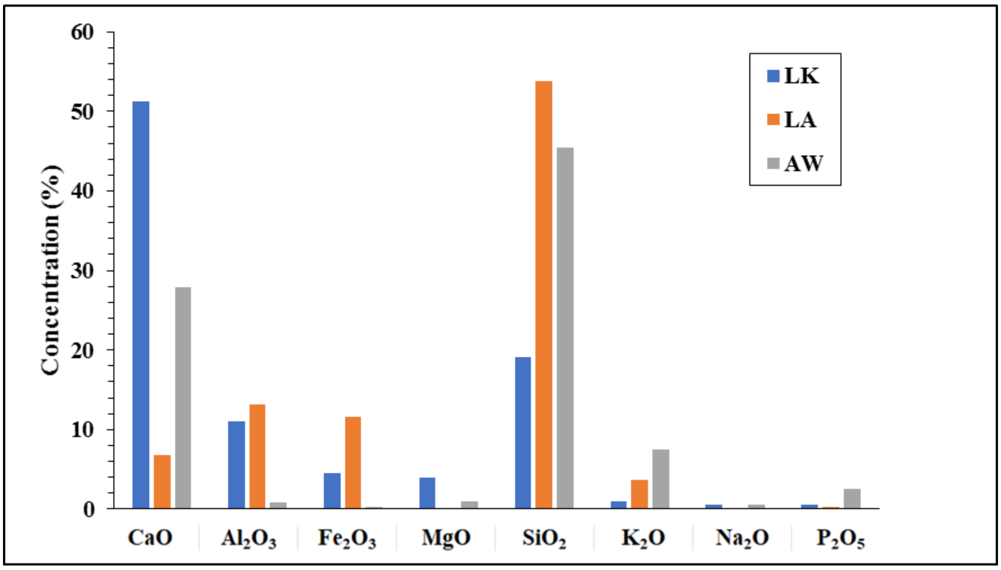

Figure 1 Results of the chemical analysis of ashes from different samples for revealing their major inorganic elements.

3.2 Gasification Characteristics of the Lignites, Acacia Waste Wood Chars, and Their Different Mixtures

A comparative analysis of the gasification profiles of the biochar fuels and their different blends as a function of temperature was performed. The results are illustrated in Figure 2 and Figure 3. The characteristic parameters of the gasification process are summarized in Table 2. It was observed that the Boudouard reaction occurred at temperatures above 600 °C. The reaction rate of Acacia waste wood was approximately 2–3-fold higher than the rates determined for the two lignites, although the maxima for the former occurred at a higher temperature (939 °C). The order of reactivity values was as follows: AW > LK > LA. Acacia waste wood exhibited the highest conversion (96.4% dry), and its cold gas efficiency reached a value of 36.3%. These variations in the gasification reactivity among the evaluated samples were attributed to the differences in the structure and chemical composition of the different fuels, considering that the experimental conditions for pyrolysis and gasification processes were identical. Previous studies [6, 12] have demonstrated that surface area and pore volume are elevated after the evolution of volatile species. However, the low concentration of alkali metals K and Na (known to indicate catalytic activity [10, 15-17]) in all samples did not allow for determining a correlation of these parameters with the reaction rate. Nevertheless, the amounts of alkaline earth metals (Ca and Mg) in Acacia waste wood and Kardia lignite were higher than the corresponding amounts in Ahlada lignite, which most probably affected the gasification rate positively. In addition, the lower rate and reactivity observed in the case of Ahlada lignite char could be attributed to its higher contents of silicon and aluminum inhibitors and also to its high ash content (47.2%), which served as physical barriers to the gasifying agent.

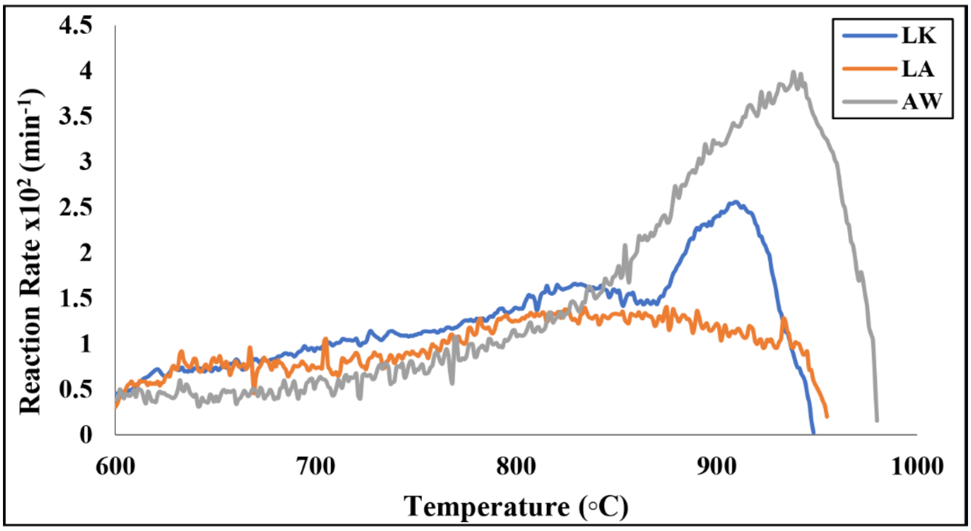

Figure 2 Gasification profiles of the evaluated biochar fuels (RSD = 0.4%–0.6%).

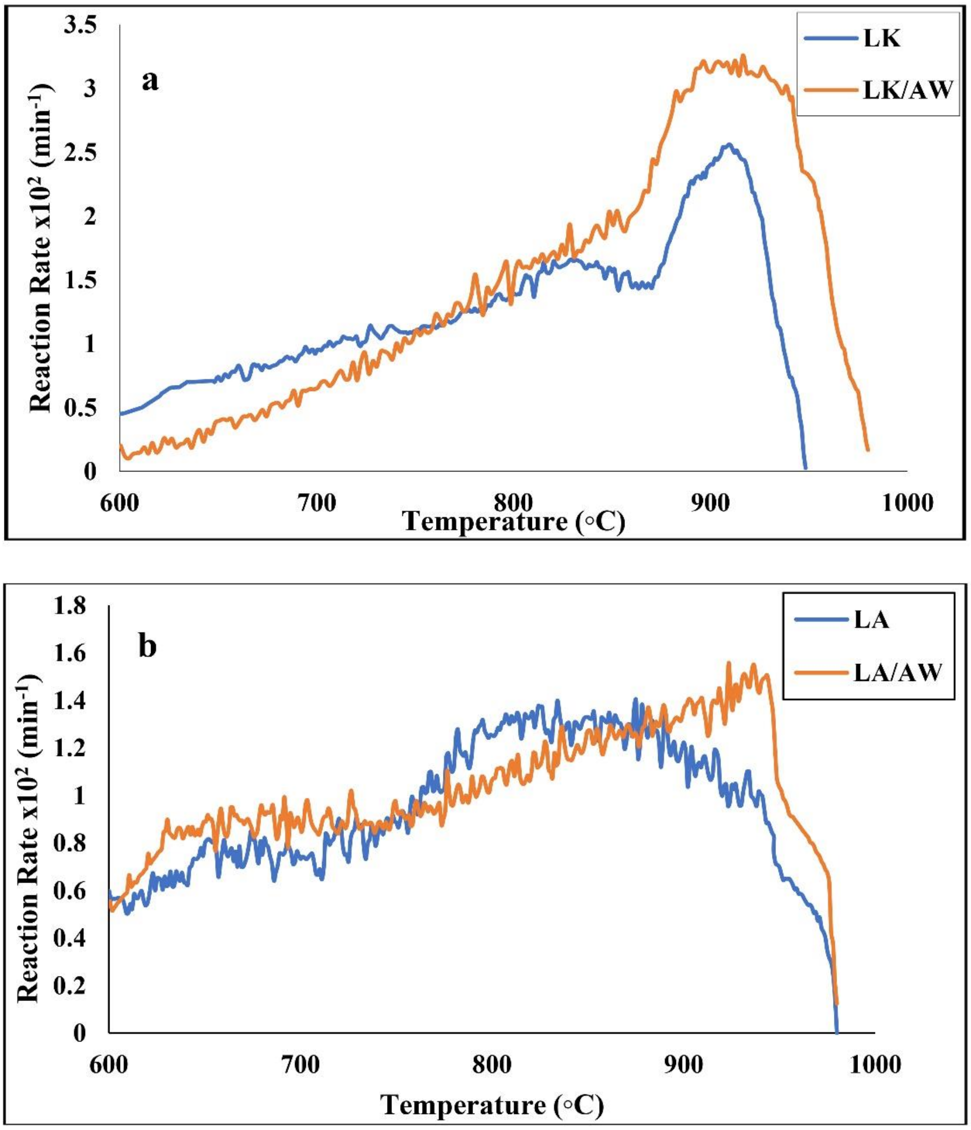

Figure 3 Gasification profiles of the blends of (a) Kardia lignite and (b) Ahlada lignite (RSD = 0.4%–0.9%).

Table 2 Gasification parameters of the evaluated biochar fuels and blends (dry basis).

Sample	**T_i (°C)**	**T_{max} (°C)**	**R_{max}x10^2 (min^{-1})**	**R_fx10^4 (min^{-1}/°C)**	**α (%)**	**CGE (%)**
LK	650	913	2.62	0.29	51.6	36.9
LA	600	859	1.37	0.16	43.4	33.5
AW	670	939	4.46	0.47	96.4	36.3
LK/AW	650	914	3.56	0.39	64.7	32.2
LA/AW	650	938	1.56	0.17	57.1	28.9

In regard to the different blends of the two lignites with the biomass material, it was observed that the component fuels reacted quite independently (refer to Figure 3), as evidenced by the peak position of the LK/AW mixture being practically the same as that of the LK while that of the

LA/AW mixture being the same as that of AW. This implied that when the reactions of component fuels occurred at different temperature regimes, mutual interactions were less favored. Therefore, in the case of the LK/AW mixture, the additivity effects dominated. However, in the case of the LA/AW mixture, the reaction rate was lower than the mean of the component rates, for the reasons previously explained. In addition, as visible in Table 2, when the char of either of the two lignites was blended with Acacia waste wood char, followed by gasification, the final conversion was increased up to 32%.

3.3 Influence of External Catalysts on the Gasification Performance of the Lignites, Acacia Waste Wood Chars, and Their Different Mixtures

The influence of the catalysts CaO and K_2CO_3 used at a concentration of 30% on the characteristic gasification parameters and the conversion of the two lignites, Acacia waste wood, and their different mixtures was evaluated. The results are presented in Table 3. It was observed that the presence of catalysts shifted the peak inflection temperature to a much lower value of up to 120 °C, particularly when K_2CO_3 was used as the catalyst. This effect would be quite beneficial in industrial appliance applications as it would reduce the slagging/fouling phenomena and the overall process cost [10, 16]. Furthermore, the addition of CaO enhanced the reaction rate and reactivity for all fuels, while the addition of K_2CO_3 slightly decreased these parameters (Figure 4). According to the mechanism of gasification, the number of active sites is increased, and the physicochemical properties are enhanced in the presence of catalysts, which leads to rapid conversion. In addition, a catalyst could alter the identity and reactivity of the surface intermediates and/or reduce the reaction temperature [10, 20]. Therefore, the different behaviors exhibited by CaO and K_2CO_3 could be attributed to the combined effects of the above-stated reasons. The small decline in the reaction rate in the presence of K_2CO_3 could be attributed to the following: (a) vaporization of K at high temperatures, (b) irreversible reactions between K and the ash components present in the sample, and (c) inactive K species formed, particularly for the coal chars with high Si and Al contents [1, 6, 17]. Table 3 also revealed that the 30% catalyst loading increased the conversion of the lignites to carbon monoxide gas by 35%–40% and that of Acacia waste wood to 100%. In terms of the gasification reactivity and conversion, CaO exhibited better catalytic performance, while the order of the overall catalytic performance was as follows: AW > LK > LA. Moreover, CaO presented an additional advantage over K_2CO_3 as the former is thermally stable and has a higher melting point (at >1300 °C), which reduces the operational issues encountered in boilers [10].

Table 3 The gasification parameters for all biochar fuels and blends in the presence of catalysts (dry basis).

Sample	**Catalyst**	**T_i (°C)**	**T_{max} (°C)**	**R_{max}x10^2 (min^{-1})**	**R_fx10^4 (min^{-1}/°C)**	**α (%)**
LK	CaO	648	894	4.01	0.44	72.0
	K_2CO_3	600	858	1.93	0.22	63.5
LA	CaO	600	898	2.36	0.26	58.4
	K_2CO_3	600	870	1.24	0.14	51.5
AW	CaO	700	898	5.17	0.57	100

	K_2CO_3	600	821	4.10	0.49	100
LK/AW	CaO	600	900	4.02	0.45	89.6
LA/AW	CaO	600	894	3.11	0.44	71.7

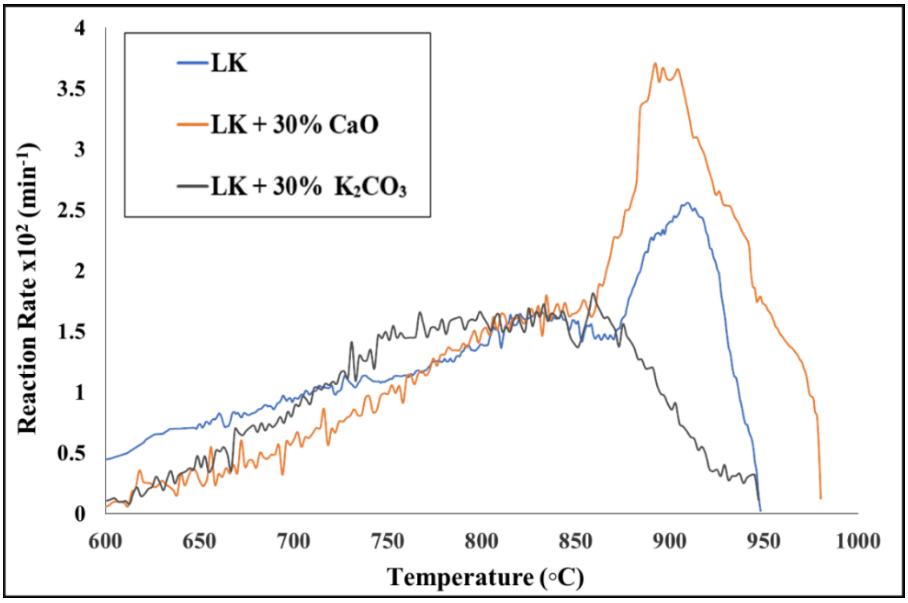

Figure 4 The DTG curves for the Kardia lignite in the absence/presence of catalysts (RSD = 0.4%–0.7%).

After obtaining the above results, the effects of the CaO catalyst on the performances of the different blends of the two lignites with the biomass material were investigated further. As visible in Table 3, the addition of 30% CaO to the blends allowed the gasification process to occur at lower temperatures compared to the temperatures at which the process occurred for the lignites. The reaction rate and reactivity were doubled, while the conversion was elevated by 74% for the Kardia lignite and 65% for the Ahlada lignite, and the corresponding final values were 89.6% (dry) and 71.7% (dry), respectively. Therefore, it was inferred that using equal amounts of Kardia lignite and Acacia waste wood in a blend would allow achieving the complete conversion of the organic matter into carbon monoxide gas during the gasification process.

The next step in the investigation was to determine the influence of a lower amount of catalyst on the conversion and the process characteristics of the different LK-AW blends. As visible in Table 4, even a lower amount of catalyst (10%) intensified the gasification of Acacia waste wood fuel; the reaction rate and reactivity were increased, and a complete conversion was achieved. The effects of catalyst loading on the performances of the different blends are illustrated in Figure 5 and Table 4. A comparison of the blend of Kardia lignite with Acacia waste wood material with or without the addition of CaO catalyst revealed that the catalyst loading of less than 30% enhanced the reactivity and conversion, although only to a small extent. On a dry-ash-free basis, at the CaO loadings of 10% and 20%, the conversion values reached 76.5% and 93.8%, respectively. Interestingly, the addition of a lower amount of CaO catalyst lowered the peak inflection temperature of the process (by 33 °C), which implied that this inorganic compound influenced the temperature sensitivity of the reaction in the presence of carbon dioxide. These findings were consistent with the previously reported data on the catalytic gasification of lignite and biomass fuels under a carbon dioxide atmosphere [6, 17, 20, 21].

Table 4 The gasification parameters of AW and LK/AW in the presence of the CaO catalyst (dry basis).

Sample	Catalyst loading (%)	T_i (°C)	T_{max} (°C)	R_{max}x10^2 (min^{-1})	R_fx10^4 (min^{-1}/°C)	α (%)
AW	-	670	939	4.46	0.47	96.4
	10	650	939	4.57	0.49	100
	20	650	937	4.86	0.52	100
	30	700	898	5.17	0.57	100
LK/AW	-	650	914	3.56	0.39	64.7
	10	600	881	3.33	0.38	65.8
	20	600	893	4.0	0.44	80.7
	30	600	900	4.02	0.45	89.6

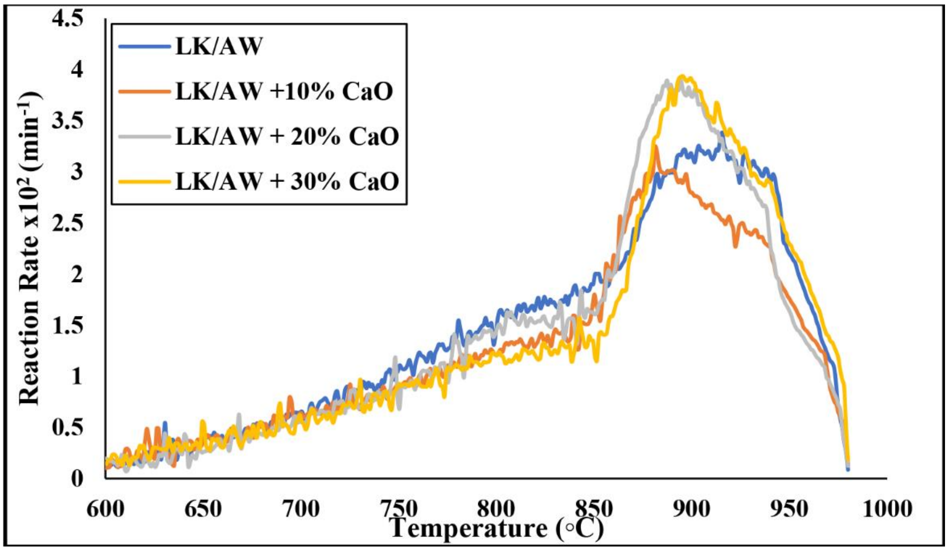

Figure 5. Effect of catalyst loading on Kardia lignite blends (RSD = 0.6%–1.0%).

4. Conclusions

Thermal treatment of lignites and biomass material prior to the gasification process produced upgraded and cleaner fuels compared to the raw materials. The reaction rate of Acacia waste wood was observed to be 2–3-fold higher than that of lignites. In addition, the conversion of Acacia waste wood at 1000 °C reached a value of 96.4% (dry basis), while that of lignites varied between 43.4% and 51.6%. The peak inflection temperatures occurred between 859 °C and 939 °C. The reactivity values of the lignite/biomass blends were higher than those recorded for the lignites, with the final conversion of the former observed to be increased by approximately 30% compared to the lignites.

Furthermore, after impregnating individual biochars with 30% CaO or K_2CO_3, the gasification process was observed to occur at lower temperatures, and the conversion was increased by 35%–40% for the lignites and the corresponding value for Acacia waste wood reached 100%. The CaO catalyst exhibited a better performance overall. In addition, the blends containing equal amounts of the lignite and Acacia waste wood, with the addition of 30% CaO, resulted in a conversion [to carbon monoxide gas] value of 89.6% and 71.7% for Kardia lignite and Ahlada lignite, respectively. Moreover, at a CaO catalyst loading of less than 30%, the reactivity and conversion values for the biochars were enhanced. Furthermore, for the Kardia lignite/Acacia waste wood blends, it was observed that the lower the amount of the catalyst added, the lower the peak inflection temperature. At 20% CaO loading, the conversion of the organic matter reached 93.8%.

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Author Contributions

D. Vamvuka: concept, writing, evaluation; k. Loulasi: experiments.

Competing Interests

The authors have declared that no competing interests exist.

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