

Thermal Behavior of Coal and Biomass Blends in Inert and Oxidizing Gaseous Environments

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ABSTRACT

Oxy-fuel combustion and gasification (pre-combustion) may have potential for capturing carbon dioxide at lower costs for power generation. Oxy-co-firing and co-gasifying coal with biomass could further reduce effective CO₂ emissions and utilize renewable energy resources. A key feature of these two approaches is that they process fuel in concentrated CO₂ or O₂/CO₂ instead of N₂ or O₂/N₂. Accurate predictive models of these processes using blends of coal and biomass can be used in process simulation and could aid in the development and implementation of these technologies. To develop these accurate predictive models, it is important to understand the conversion routes and thermal behavior of these fuels in appropriate gas environments. The objectives of this study are to investigate the impact of inert and oxidative gaseous environments on thermal behavior and reactivity of coal and biomass blends and to study the effect of biomass percentage on coal/biomass blend co-utilization. Fuel samples included a Powder River Basin (PRB) sub-bituminous coal, yellow pine wood sawdust pellets, and mixtures of 10 and 20 weight percent wood in coal. The samples were tested under N₂, CO₂, and 10% O₂ in CO₂ by volume using a non-isothermal thermogravimetric method for temperatures up to 1000°C. Fuel weight losses of both coal and wood are essentially the same in CO₂ as in N₂ in the low temperature range, but higher in 10% O₂ in CO₂ compared to N₂ and CO₂. However, total weight losses at 1000°C under CO₂ and 10% O₂ in CO₂ are similar and higher than in N₂ due to char gasification by the CO₂ and combustion by O₂. The char combustion in 10% O₂ in CO₂ takes place at lower temperature than char gasification in CO₂. Coal and wood blends have higher reactivity compared to coal alone in the lower temperature range due to the high volatile matter content of wood. Interactions of wood and coal in these gas environments and blend percentage are discussed.

Keywords: Pyrolysis; Gasification; Combustion; Coal-Biomass Blends

1. Introduction

Coal is the dominant energy resource for electricity generation in the US because coal is abundant and less expensive than other options. The combustion of coal to generate electricity emits pollutants such as gaseous oxides of sulfur (SO₂) and nitrogen (NO_x) and the greenhouse gas CO₂. Co-firing technology simultaneously fires coals with biomass in a coal-fired boiler. It generates “green” power by utilizing renewable energy resources and reduces coal CO₂ emission since biomass is renewable and carbon neutral [1,2]. In addition, most biomass has little or no sulfur or nitrogen, therefore co-firing could lower SO₂ and NO_x levels. These co-firing advantages were demonstrated in most of the co-firing tests in Europe and the United States (depending on biomass used) [3-6]. The tests were at low biomass fraction (typically 20% or less) [3,4], and wood was the predominant biomass compo-

nent [7,8].

Oxy-fuel combustion and integrated gasification combined cycle (IGCC) with CO₂ capture (pre-combustion) technologies may have potential for capturing the greenhouse gas carbon dioxide at lower costs. In both processes oxygen is used in a coal boiler or gasifier instead of air and produces mainly CO₂ and water in the flue gas from oxy-fuel [9] and concentrated carbon dioxide in syngas from gasification [10,11], and thus has a benefit for CO₂ capture. Oxy-co-firing and co-gasification of coal with biomass could further reduce effective CO₂ emissions and utilize renewable energy resources.

Oxy-fuel combustion burns coal in O₂/CO₂ environments instead of O₂/N₂ as in conventional co-firing. Similarly, the gasification process is also in an O₂/CO₂ environment in the gasifier. Pilot and laboratory scale experiments have revealed some differences in oxy-fuel combustion and conventional air combustion, including effects such as delayed ignition and reduced flame tem-

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perature [12,13]. Higher heat capacity of CO₂ and lower oxygen diffusion rate in CO₂ compared to N₂ contribute to these effects. In addition, the oxy-fuel combustion process is affected by fuel properties [14], and differences between biomass and coal are expected to have additional significant impact.

To accelerate the deployment of commercial oxy-fuel and gasification power plants, the development of advanced and validated simulation tools such as computational fluid dynamics (CFD) models is a potentially effective approach. These models need to account for a measure of fuel flexibility. This fuel flexibility should not only include the wide range of variability expected in coal compositions but should also include the possibility of coal and biomass co-utilization. The development of improved and validated CFD models requires accurate characterization of the thermal behavior of different fuels in different gas environments. Experimental studies are necessary to understand the mechanisms of the combustion and gasification processes and to obtain experimental data needed to validate the models. The objectives of this study are to investigate the impact of inert and oxidative gaseous environments on thermal behavior and reactivity of coal, biomass and blends and to study the effect of biomass percentage on coal/biomass blend thermal behavior.

2. Materials and Experiments

2.1. Materials

A Powder River Basin (PRB) sub-bituminous coal and a yellow pine wood sawdust pelletized material were selected for this study. These feedstocks were obtained from the US Department of Energy's National Carbon Capture Center (NCCC) managed by Southern Company. The received PRB coal was already ground and dried, and is a subsample of the material that is fed in their operating plant. The wood pellets are cylindrical in shape with a diameter of 8 mm and 32 mm long. They were pro-

duced from southern yellow pine with less than 1 wt% bark (no added chemicals) by Green Circle Bio Energy, Inc. The proximate, ultimate and ash mineral analyses results listed in **Table 1** were provided by Southern Company. Wood pellet samples were ground using a high speed rotary mill. Both coal and the ground wood were sifted with a sifter and finally dried in an oven. The particle size fraction of 100 to 300 μm was used for all experiments. The proximate, ultimate and ash composition analyses of prepared samples were obtained. The analysis results of received and prepared samples are similar (data not shown) so the prepared samples are expected to be representative of the received materials. The wood has dramatically different properties compared to the coal. It has high volatility, low contents of S and N, low heating value and high potassium.

2.2. Experiments

The reactivity and thermal behavior of PRB coal, wood, and blends of 10% and 20% (wt.) wood in coal were studied in inert gas N₂ and oxidizing gases of CO₂ and 10% O₂ in CO₂ (O₂:CO₂ = 1:9 by volume). The tests were conducted by a using non-isothermal method in a PerkinElmer Pyris 1 thermogravimetric analyzer (TGA). Samples of approximately 10 mg fuel were heated from room temperature to 100°C at 20°C/min and held at 100°C for 20 min; the samples then heated to 1000°C at 20°C/min. The same gas was used for purge and process gas flow with a total flow rate of approximately 125 ml/min. The experiments were performed in triplicate (quadruplicate or more for 10% O₂ in CO₂, particularly for samples with wood) to assess their reproducibility.

The TGA thermograms recorded percent sample weight as a function of measured sample temperature (or time). The first derivative of the thermal curves (DTG) was calculated for each thermogram to facilitate identification of multi-step processes. The thermal behavior has been characterized by calculating the percent weight loss, ΔW, for

Table 1. Proximate, ultimate and ash composition of powder river basin (PRB) sub-bituminous coal and wood*.

	Proximate analysis (% dry basis)			Ultimate analysis (% dry basis)					HHV (Btu/lb)				
	volatile matter	fixed carbon	ash	C	H	N	S	O(diff)	(dry basis)				
PRB coal	40.83	50.34	8.83	67.24	4.23	1.53	0.38	17.79	11,439				
Wood pellet	85.19	13.40	1.42	53.20	6.24	0.12	0.02	39.00	8839				
Ash mineral analysis (oxides and ignited % wt.)													
	Aluminum	Barium	Calcium	Iron	Magnesium	Manganese	Phosphorous	Potassium	Silicon	Sodium	Strontium	Sulfur	Titanium
PRB coal	16.00	0.52	19.24	5.52	4.68	-	0.96	0.75	38.71	1.22	0.23	10.69	1.08
Wood pellet	13.80	0.21	21.20	4.15	6.12	2.57	1.74	4.06	37.80	1.07	0.30	5.50	0.77

*provided by Southern Company.

each thermal process (dehydration, devolatilization/pyrolysis, gasification and/or combustion) and the temperature of maximum rate of weight loss (T_{max}) for each well-separated weight loss feature, as well as the final percentage of residue remaining at end of each experiment. The percent final residue or solid yield (dry basis) is calculated as $m_f/m_{100} * 100$, where m_f and m_{100} are the weights of the feedstock sample after heating to 1000°C and after drying at 100°C, respectively.

Interactions of wood and coal in these gas environments and blend percentages are checked by comparison of experimental thermal curves and calculated weight loss profiles of blends based on weight loss of parent fuels and the ratio of blending. The calculated weight loss profile for the blend is given by

$W_{blend,i} = r_{wood} * W_{wood,i} + (1 - r_{wood}) * W_{coal,i}$, where r_{wood} is the weight fraction of wood in the blends, and $W_{wood,i}$ and $W_{coal,i}$ are the percentage sample weight of wood and coal, respectively, at temperature i . If the experimental

weight loss was not significantly different from the calculated one, this indicates that synergistic effects between coal and wood are absent at the selected experiment conditions. In this case, the weight loss of blends can be predicted by a linear relation of parent fuel weight loss properties and the percentage wood in the blends.

3. Results and Discussion

3.1. Thermal Events, Thermal Behavior and Reactivity of Coal and Wood in N_2 , CO_2 and 10% O_2 in CO_2

Figure 1 shows curves of the weight loss (TG) and weight loss rates (DTG) of the wood and coal samples as function of temperature in inert (N_2) and oxidizing gases of CO_2 and 10% O_2 in CO_2 . The weight loss below 101°C is attributed to moisture loss from the samples in all gases. In N_2 , devolatilization/pyrolysis took place as expected.

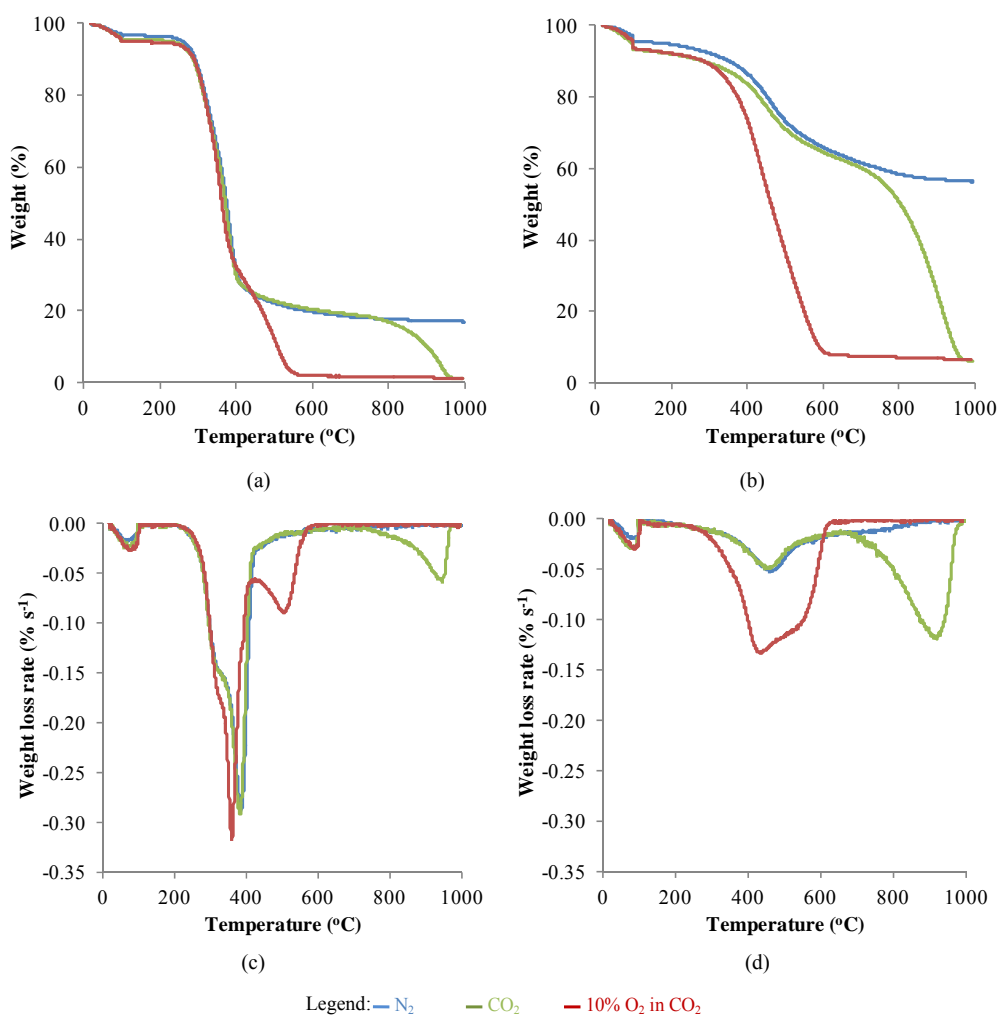


Figure 1. The sample weight percent and derivative weight loss curves (TG and DTG) of yellow pine wood ((a) and (c)) and PRB coal ((b) and (d)) in N_2 , CO_2 , and 10% O_2 in CO_2 .

The weight loss is due to release of gases (such as CO, H₂, CH₄, CO₂ and H₂O) and tars as volatile matter (VM) by thermal rupture of chemical bonds in the fuel. The residual solid char is essentially carbon (fixed carbon, FC) with mineral matter and some of the nitrogen and sulfur [15]. The DTG curve for coal pyrolysis has a broad peak with higher temperature $T_{\max} = 463^{\circ}\text{C} \pm 2^{\circ}\text{C}$ and lower maximum weight loss rate $R_{\max} = 5.2 \times 10^{-2}\% \text{ s}^{-1}$ compared to wood which has a sharp peak in the derivative with lower temperature of maximum rate of weight loss, $T_{\max} = 385^{\circ}\text{C} \pm 2^{\circ}\text{C}$, and higher maximum loss rate $R_{\max} = 2.9 \times 10^{-1}\% \text{ s}^{-1}$. As expected, coal pyrolysis gives higher percent char yield $Y_{\text{char}} = 56.4 \pm 0.2$ (wet basis) and lower volatiles $Y_{\text{vol}} = 39.0 \pm 0.3$ (wet basis) compared to wood $Y_{\text{char}} = 17.0 \pm 0.2$ and $Y_{\text{vol}} = 79.6 \pm 0.2$ (wet basis).

In CO₂, total weight losses at 1000°C for coal and wood are higher than in N₂ due to the combination of CO₂ char gasification and devolatilization (**Figure 1**). These two process steps are clearly seen in the DTG curves. Devolatilization occurs similarly to that seen in N₂ and is followed by char gasification at the higher temperatures. The char gasification is mainly via the Boudouard reaction $\text{C}(\text{s}) + \text{CO}_2 \rightleftharpoons 2\text{CO}$ [15]. It takes place at lower temperature, $T_{\max} = 918^{\circ}\text{C} \pm 4^{\circ}\text{C}$, and at higher rates $R_{\max} = 1.2 \times 10^{-1}\% \text{ s}^{-1}$ for coal compared to wood $T_{\max} = 946^{\circ}\text{C} \pm 5^{\circ}\text{C}$ and $R_{\max} = 5.6 \times 10^{-2}\% \text{ s}^{-1}$. This is in agreement with the studies by Rathnam *et al.* [16], who used coals in a drop tube reactor (DTR) at 1400°C and Al-Markhadme and Scheffknecht [17] who examined coals in an entrained flow reactor (EFR) at temperatures from 700°C to 1150°C. However, the opposite results were obtained in a DTR for coals at 1300°C by Borrego and Alvarez [18] and for biomass at 950°C by Borrego *et al.* [19]; they attributed this to CO₂ participation in cross-linking reactions at the surface of the devolatilizing particles.

In 10% O₂ in CO₂, total weight losses at 1000°C for coal and wood are the same as in CO₂ and higher than in N₂ due to mainly char combustion (**Figure 1**). The fuel thermal decomposition process has two main steps based on DTG curves. Devolatilization and volatile combustion are followed by char combustion, which are clearly shown in two main regions of weight loss for wood but overlapped in one broad feature for coal. The weight loss in the char combustion is similar to that observed in the char gasification, which is clearly seen by comparing the thermal curves in CO₂ and in the mixture of oxygen and CO₂ for wood. In the presence of oxygen, devolatilization rates are the same as with N₂ at lower temperatures and then faster than in N₂ and CO₂ environments at a slightly higher temperatures. This is likely due to volatiles combustion. Above a certain temperature, once volatiles are released from the solid, these compounds undergo oxidation within the gas film surrounding the particle and result in particle temperature increases.

3.2. Effect of Biomass Percentage on the Reactivity and Thermal Behavior of Coal/Biomass Blends

Figure 2 shows TG and DTG curves of the coal, wood, 10% and 20% wood in coal blends as function of temperature in oxidizing gases of CO₂ (a) and 10% O₂ in CO₂ (b). TG and DTG curves of the blends in N₂ are the same as in CO₂ except without the gasification event. The weight losses of the blends are higher than coal alone in both the devolatilization and char gasification regions but same with coal in combustion region. Devolatilization occurs over a wider temperature range in CO₂ than in 10% O₂ in CO₂. The dried blends have two different regions of weight loss. In the lower temperature range ($T < 415^{\circ}\text{C}$ in N₂ and CO₂ and $T < 390^{\circ}\text{C}$ in 10% O₂ in CO₂), the trends of weight loss rates for blends are similar to that of wood. In the higher temperature region, however, they are quite close to coal since wood has low fixed carbon, and there is a low ratio of wood in these blends. Similarly, the blends have temperatures for maximum rate of weight loss (T_{\max}) close to that of wood in the lower temperature region, and comparable to coal in the higher temperature region (**Table 2**). The weight loss rates of blends are higher than coal alone in all three gases in the lower temperature region (**Figure 2**) due to high volatile matter (VM) (**Table 1**) and reactivity of wood. In CO₂, the weight loss rate of the 10% wood blend at $T_{\max} = 385^{\circ}\text{C}$ ($6.7 \times 10^{-2}\% \text{ s}^{-1}$) is 2.7 times higher than for coal at the same temperature ($2.5 \times 10^{-2}\% \text{ s}^{-1}$). However, in 10% O₂, the rate at $T_{\max} = 374^{\circ}\text{C}$ ($7.7 \times 10^{-2}\% \text{ s}^{-1}$) is only 1.3 times higher than coal ($6.0 \times 10^{-2}\% \text{ s}^{-1}$) at that temperature. In addition, the 20% wood blends have higher weight loss rates than the 10% wood blends in N₂ and CO₂ but both blends have similar weight loss rates in 10% O₂ in CO₂.

As described in Section 2.2, the final residues of blends after heating to 1000°C were calculated based on final residues (dry basis) of parent fuels and ratio of blending. These are compared with experimental results in **Table 3**. There is no significant difference between experimental and calculated final residues of blends. As discussed above, the wood affects the blends more in the lower temperature ranges. So, a similar comparison of calculated weight loss to observed experimental loss was performed in the low temperature range from 100°C to 415°C. As can be seen in **Figure 3**, there are no significant differences between the weight losses from experiments and calculation in N₂ and CO₂ but the two values do appear to be slightly different in 10% O₂ in CO₂. However, these differences are quite small and may not be statistically significant.

As described in Section 2.2, TG curves of blends were calculated based on weight loss profiles of the parent fuels

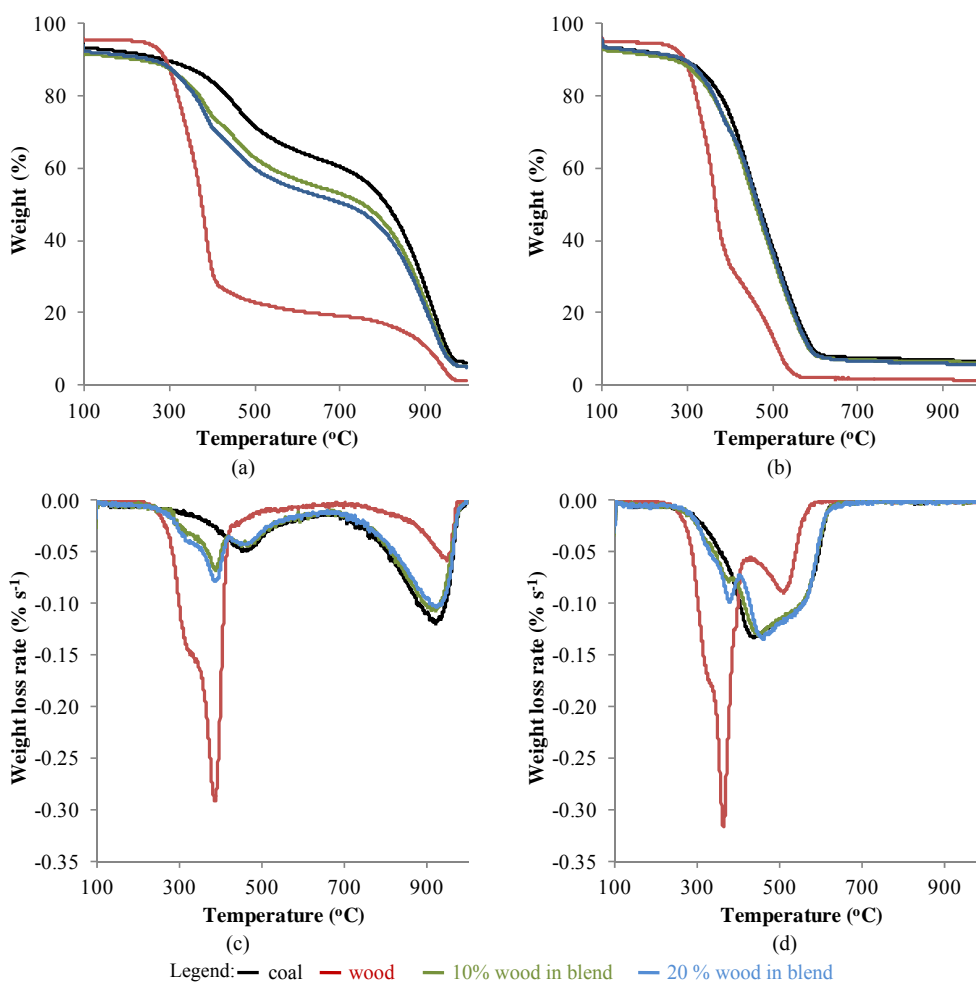


Figure 2. The percent sample weight and derivative weight loss curves (TG and DTG) of PRB coal, yellow pine wood, 10 wt% and 20 wt% wood in blend in CO₂ (a) and 10% O₂ in CO₂ (b).

Table 2. Maximum temperatures of PRB coal, yellow pine wood and their blends in N₂, CO₂, and 10% O₂ in CO₂.

Gas	Sample	Maximum temperature (°C)		
N ₂		devolatilization		
	PRB coal	463 ± 2		
	10 wt% wood in blend	380 ± 4		
	20 wt% wood in blend	379 ± 2		
	Wood pellet	385 ± 2		
CO ₂		devolatilization		
	PRB coal	457 ± 7		
	10 wt% wood in blend	385 ± 4		
	20 wt% wood in blend	382 ± 3		
	Wood pellet	383 ± 1		
			char gasification	
	PRB coal		918 ± 4	
	10 wt% wood in blend		914 ± 6	
20 wt% wood in blend		913 ± 13		
Wood pellet		946 ± 5		
10% O ₂ /CO ₂		devolatilization/ volatile combustion		
	PRB coal	432 ± 4		
	10 wt% wood in blend	440 ± 11		
	20 wt% wood in blend	453 ± 7		
	Wood pellet	369 ± 13		
		char combustion		
			510 ± 12	

Table 3. Comparison of final residues (dry base) of PRB coal, yellow pine wood and their blends after heating to 1000°C in N₂, CO₂, and 10% O₂ in CO₂. Calculated values are based on parent fuel values and blending ratios as described above in section 2.2.

Samples	Final Residue (%)		
	N ₂	CO ₂	10% O ₂ /CO ₂
PRB coal	59.1 ± 0.2	6.6 ± 0.3	7.1 ± 0.1
Wood pellet	17.6 ± 0.2	1.3 ± 0.1	1.2 ± 0.2
10 wt% wood in blend	54.3 ± 1.1	5.5 ± 0.3	6.7 ± 0.5
20 wt% wood in blend	48.0 ± 2.3	5.3 ± 0.5	5.1 ± 0.5
Calculation based on parent residues and ratio of blend			
10 wt% wood in blend	55.0	6.1	6.5
20 wt% wood in blend	50.8	5.5	5.9

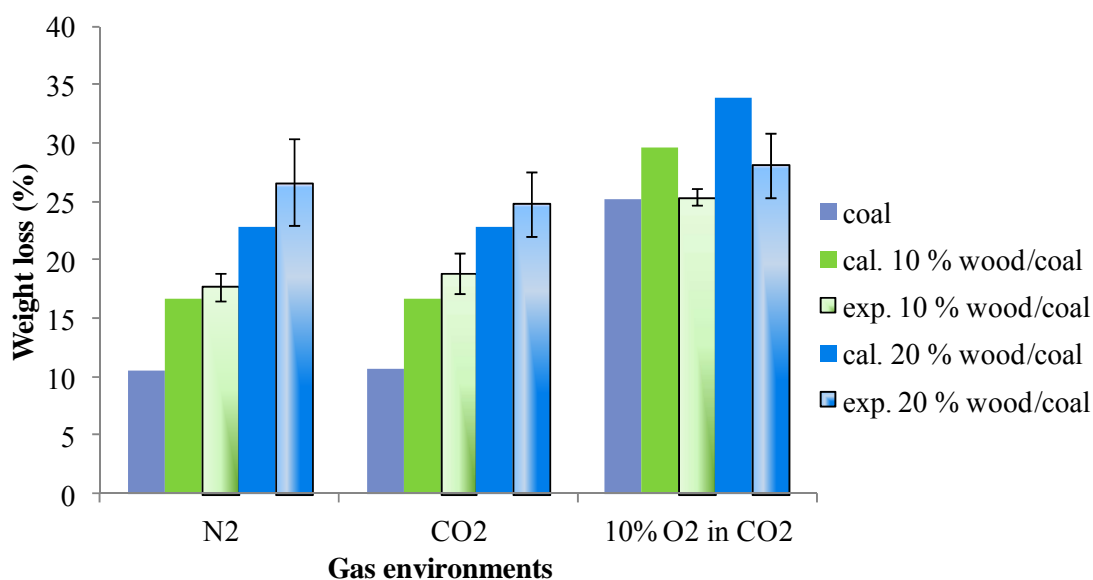


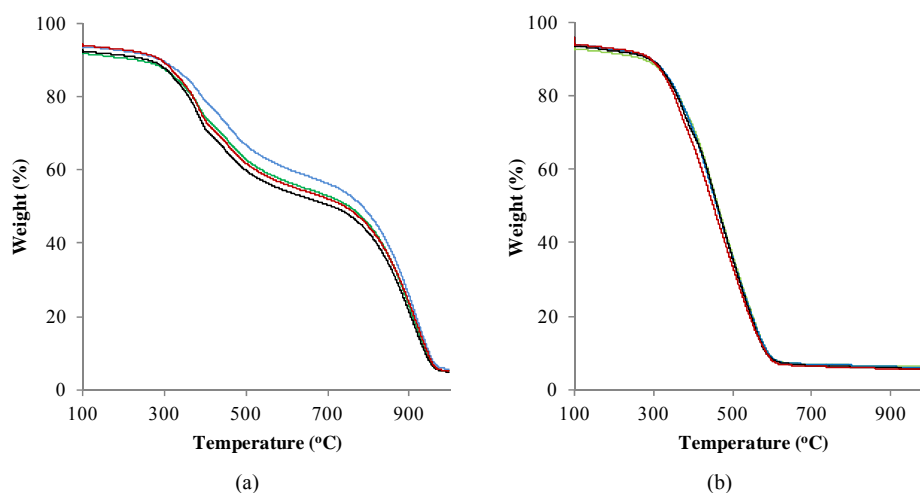
Figure 3. Comparison of experimental weight loss (dry basis) of coal and wood blends in N₂, CO₂, 10% O₂ in CO₂ from experiments and calculated weight loss based on parent fuel thermograms and ratio of blends in 100°C - 415°C.

and the ratio of fuel blending for N₂, CO₂ and 10% O₂ in CO₂ environments. Comparison of these calculated curves with the experimental TG curves showed no significant differences (Figure 4. Note, N₂ data not shown). Overall, there appear to be no significant interaction observable from the weight loss profiles alone between coal and biomass in the solid phase during co-pyrolysis, co-gasification, and co-combustion in this study. These may be explained by the low ratio of wood char in the blend char. Based on the char yields (final residues) of coal and wood from the experimental results in N₂, the ratio of wood char contribution to the total blend char for 10 and 20 wt% wood blends is estimated to be 3.2% and 6.9%, respectively. This result is in agreement with the results obtained by Biagini *et al.* [20], Vuthaluru [21], and Moghtaderi *et al.* [22] on co-pyrolysis and Gil *et al.* [23] on co-combustion using TGA. For co-pyrolysis the same results are obtained using a drop tube furnace [22]. To shed

light on the issue of possible reaction between coal and biomass or synergetic effect of biomass on coal, further study is needed because of the inherent heterogeneity of wood and coal, the small sample size, and the small number of replicate determinations used in the present study.

4. Conclusion

Oxy-co-firing (combining biomass co-firing and oxy-fuel technologies) and co-gasification of coal and biomass together could further reduce effective CO₂ emissions and utilize renewable energy resources. These processes gasify and combust fuels in concentrated CO₂ and O₂/CO₂ instead of N₂ and O₂/N₂. The present study investigates the impact of biomass percentage on the thermal behavior of coal and biomass blends in inert (N₂) and oxidizing gases (CO₂ and 10% O₂/CO₂). The PRB sub-bituminous coal, yellow pine wood pellets, and blends of coal with



Legends: — exp. 10 % wood in blend — cal. 10 % wood in blend — exp. 20 % wood in blend — cal. 20 % wood in blend

Figure 4. Comparison of TG thermograms of coal and wood blends from experiments and calculated based on parent fuel thermograms and ratio of blends, in CO₂ (a) and in 10% O₂/CO₂ (b).

up to 20 weight percent of this wood are being studied using thermogravimetry for temperatures up to 1000°C. Different thermal events of pyrolysis, gasification and combustion takes place in different temperature ranges in these three gaseous environments. In the presence of oxygen, devolatilization rates are faster than in N₂ and CO₂ environments due to the volatiles combustion. The total fuel weight losses at 1000°C for coal, wood and blends were higher in CO₂ than in N₂ due to CO₂ char gasification in addition to the devolatilization. Char combustion in 10% O₂ in CO₂ takes place at lower temperature than char gasification in CO₂. The blending of wood increases fuel weight loss rate in the lower temperature range in all gases due to the higher volatile matter content and narrower temperature range of devolatilization for wood compared to coal. Thermal processes for coal/wood blends can be divided into two reaction regions by temperature. In the lower temperature range, the blend thermal behavior is more like that of the biomass, and in the higher temperature range, it is similar to coal. There appear to be no significant interactions between coal and biomass observable by thermal degradation weight loss profiles for the blends with low wood percentage in N₂, CO₂ and 10% O₂/CO₂.

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