



# Mineral matter interactions during co-pyrolysis of coal and biomass and their impact on intrinsic char co-gasification reactivity



Naoko Ellis<sup>a,\*</sup>, Mohammad S. Masnadi<sup>a,\*</sup>, Daniel G. Roberts<sup>b</sup>, Mark A. Kochanek<sup>b</sup>, Alexander Y. Ilyushechkin<sup>b</sup>

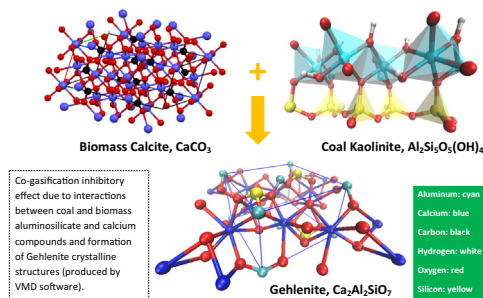
<sup>a</sup> Clean Energy Research Centre, Department of Chemical & Biological Engineering, University of British Columbia, Vancouver, BC V6T 1Z3, Canada

<sup>b</sup> CSIRO Energy, PO Box 883, Kenmore, QLD 4069, Australia

## HIGHLIGHTS

- Biomass/coal pyrolysis effects on char co-gasification kinetics was investigated.
- Higher co-gasification rates were observed for the separate pyrolyzed samples.
- Lower co-gasification rates were observed for the combined pyrolyzed samples.
- Biomass and coal minerals interacted through co-pyrolysis and inhibited gasification.
- The XRD result confirmed formation of gehlenite crystals during combined pyrolysis.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Gasification of blends of biomass and coal can offer renewable fuels the scale and extent of deployment usually associated with fossil fuels. For significant penetration of renewables, however, co-utilization of significant quantities of biomass is required, which significantly impacts process performance. At a fundamental level, char reactivity affects many practical aspects of gasifier operation, and is complicated by the influence of blends of coal and biomass and their different behaviour during devolatilization. In this work, intrinsic gasification reaction kinetics of chars from biomass and coal mixtures with different proportions were studied: one set of chars produced separately and mixed prior to gasification; and another with chars produced from co-pyrolysis of biomass–coal blends. Lower specific and intrinsic rates were observed for the samples where the biomass and coal were pyrolyzed together than when they were pyrolyzed separately, suggesting some interaction during devolatilization that affects reactivity behaviour. XRD results showed that the catalytically-active calcium species in the biomass interacted with the aluminosilicate species in the coal mineral matter to form  $\text{Ca}_2\text{Al}_2\text{SiO}_7$  (gehlenite) crystals, which are catalytically inert. The conversion of catalytically-active Ca to catalytically-inactive Ca may have led to lower reactivity of co-pyrolyzed mixtures, highlighting the importance of understanding the type and nature of often catalytically-active species when investigating the gasification behaviour of blends of coal and biomass materials.

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\* Corresponding authors at: Chemical and Biological Engineering Building, 2360 East Mall, Vancouver, BC V6T 1Z3, Canada. Tel.: +1 604 822 1243; fax: +1 604 822 6003.

E-mail addresses: [naoko.ellis@ubc.ca](mailto:naoko.ellis@ubc.ca) (N. Ellis), [s.masnadi@gmail.com](mailto:s.masnadi@gmail.com) (M.S. Masnadi).

## 1. Introduction

There is global pressure for coal-dominated power generation to significantly reduce greenhouse gas emissions. In the US, for example, there are incentives for industries to diversify their energy sources to include a greater proportion of renewables [1]. Co-gasification of biomass and coal offers a transitional solution from energy production via fossil into renewable fuels [2], by offering a renewable energy resource access to the scale and extent of deployment usually only associated with fossil fuels.

There are clear benefits associated with co-gasification of biomass and fossil fuels. Addition of coal to biomass increases the carbon and energy density of the feedstock, and there are significant reductions in greenhouse gas emissions if the biomass is sustainably available. Coal can also be used to manage seasonal fluctuations in biomass availability, and in the context of waste gasification, reduce biomass material sent to landfill. Furthermore, the high amount of catalytically-active species usually present in biomass may enhance overall carbon gasification and tar reforming reactions. Overall, there are significant improvements possible in terms of energy diversity and security by utilizing biomass in conjunction with coal.

There are technical issues, however, associated with co-gasification of significant amounts of biomass that can impact technology choice, blending strategies, and the overall effectiveness of the co-gasification approach. These can often mean that coal-based technologies require significant modification to be able to accept large proportions of biomass, or as in many cases, new plant is required that is well-suited to the co-gasification of biomass-coal blends. For these reasons, research into the fundamental aspects of co-gasification of biomass and coal is important. The effect of interactions during pyrolysis on the reactivity of the char is one of the key aspects of this research, as accurate knowledge of char conversion kinetics are required for plant design, development of blending strategies, as well as optimization and troubleshooting.

Biomass-coal co-pyrolysis has been previously investigated and there is a wide range of outcomes in the literature suggesting that there are many underlying processes that are likely to be at play. Several authors have neither observed any significant positive effect on the resulting char reactivity [3,4], nor any evidence of interactions in the product distributions from co-pyrolysis [5]. In most of these studies, the gas flow swept the volatile products away from the devolatilizing fuels; therefore, the devolatilizing gas could not easily interact with the char, and thus the additive behaviour of the products prevailed [6]. On the other hand, synergistic interactions during co-pyrolysis of coal and biomass have also been reported [7–11]. These observed synergistic effects may be due to the presence of oxygen-containing species and the alkaline and alkaline-earth metal (AAEM) species such as sodium, potassium, and calcium originating from the biomass and affecting the reactivity of the char mixture. There are reports that at low AAEM concentrations, the biomass minerals are sequestered by secondary fuel (e.g. coal and biosolids) ash minerals (e.g. Si and Al) [4,12]; whereas, mixtures with high AAEM can lead to significant enhancement in the overall gasification rate [13,14]. Mineral matter transformation and reaction chemistry is a complex aspect of understanding the gasification behaviour of blends. Rowan et al. [15] suggests that varying the biomass content in the initial feed stock shows linearity with respect to the proximate and ultimate (elemental) analysis results of the produced chars; however, non-linearities can be seen in the tar and char yields, and in the functional group structure of the chars, as revealed by FT-IR analysis of the solid char samples. A recent paper reports on the effects of potassium volatilization and deposition, and the impact of

volatiles on the chemical structure of char of blended pyrolysis of corncob and lignite, highlighting the complexity in which co-pyrolysis may affect the char reactivity [16].

Although co-pyrolysis and co-gasification of biomass and coal have been studied extensively, there are few data that provide insights into the impact of co-pyrolysis on fundamental aspects of char co-gasification reactivity. In particular, our knowledge regarding intrinsic reactivity behaviour, which is the fundamental component of a feedstock's conversion reactivity behaviour, is limited and incomplete. While intrinsic reactivity parameters are measured under conditions usually far removed from any particular industrial process, they are key to our understanding of the gasification reaction system, and required for understanding feedstock behaviour in a range of relevant technologies. Therefore in this paper, the potential for interactions between coal and biomass during co-pyrolysis, and their effect on the intrinsic gasification reactivity of the chars, is investigated.

## 2. Materials and methods

### 2.1. Feedstocks

Pelletized pine sawdust from British Columbia, Canada and a high-volatile bituminous coal from NSW, Australia were chosen as the biomass and coal samples, respectively. Both fuels were crushed and dry sieved to  $-1.0 + 0.6$  mm prior to devolatilization. The proximate, ultimate, and ash analyses of the parent samples are presented in Table 1. All fresh samples were characterized according to ASTM D3176 and D5373 standards for carbon, hydrogen, nitrogen and sulphur (CHNS) contents; ASTM D4239 for total sulphur content; ASTM D3174 to determine ash content; ASTM D3302/D3173 for moisture content; ASTM D3175 for volatile matter and ASTM D5865 for heating value. Ultratrace by ICP mass spectrometry and XRF techniques were used for the biomass and coal ash elemental analysis, respectively.

As expected, the coal contained higher fixed carbon (50.8 wt%) than biomass (12.0 wt%); while, the biomass had much higher

**Table 1**  
Proximate, ultimate, and ash analyses of the samples used in this work.

Sample	NSW bituminous coal	BC pine sawdust
<i>Proximate analysis (wt%)</i>		
Moisture	2.8	8.0
Ash (db <sup>*</sup> )	13.9	0.4
Volatile (db)	35.3	87.6
Fixed Carbon (db)	50.8	12.0
Higher heating value (db) (MJ/kg)	28.7	20.4
<i>Ultimate analysis (wt%), daf<sup>**</sup></i>		
Carbon, C	83.3	50.1
Hydrogen, H	5.8	6.2
Nitrogen, N	1.8	0.0
Sulphur, S	1.1	0.0
Oxygen, O (diff <sup>**</sup> )	8.0	43.6
<i>Ash analysis (wt% of ash)</i>		
SiO <sub>2</sub>	47.9	45.1
Al <sub>2</sub> O <sub>3</sub>	26.5	13.3
TiO <sub>2</sub>	1.9	0.3
Fe <sub>2</sub> O <sub>3</sub>	7.5	2.3
CaO	7.9	19.7
MgO	0.6	4.6
Na <sub>2</sub> O	0.1	1.3
K <sub>2</sub> O	0.2	7.0
P <sub>2</sub> O <sub>5</sub>	1.3	1.4
SO <sub>3</sub>	6.1	2.0
Undetermined	0.0	3.0

<sup>\*</sup> daf = dry and ash free, db = dry basis.

<sup>\*\*</sup> Calculated by difference.

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