

## INFLUENCE OF COAL BLEND COMPONENT KINETICS ON ENTRAINED-FLOW GASIFICATION PERFORMANCE

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### ABSTRACT

The German research project *ibi* – Innovative Process Technology aims to utilise Central German brown coal primarily in a non-energetic way. This task involves the development of advanced mining concepts as well as a subsequent staged process chain for optimal coal utilisation. The final thermal conversion stage of such a process chain is an entrained-flow gasification unit that has to be able to process solid fuel blends consisting of components originating from previous process stages. Therefore, this paper addresses the challenge to predict the influence of differing coal blend ratios on entrained-flow gasification performance.

A simulation study incorporating a validated 1D kinetic entrained flow coal blend gasification model has been carried out with a blend of 3 different Australian coals from literature (Hla et al., 2007). The simulation results show synergy effects in terms of carbon conversion and cold gas efficiency due to different coal composition and different heterogeneous gasification kinetics of the blend components. Ternary graphs are used to visualize these different influences of each blend component on gasification process results.

### INTRODUCTION

Coal utilization research is steadily gaining in importance as oil depletion and gas price increase will continue during the next decades. On one hand, coal is a necessary source of energy in the transition phase towards renewable energy supply. On the other hand, the non-energetic utilization of coal represents an important supply of carbon for the chemical industry to produce base chemicals like methanol or propylene (Pardemann & Meyer, 2011). In particular coal gasification will be an indispensable technology for the carbon based chemical industry to cope with future base chemical market demands. Therefore, further development of gasification plants which are less cost intensive and with lower CO<sub>2</sub> emissions is necessary. This objective is realizable through research activities targeted at increasing the efficiency of gasification processes.

Experimental verification has shown that fuel blending represents a promising approach to achieve higher efficiency in gasification and combustion (Shen et al., 2009; Xu et al., 2011; Feroso et al. 2009; Cousins et al., 2008; Seo et al., 2010; Lee et al., 2010; Pan et al., 2000). For many years, coal blending and coal-biomass blending are applied in combustion and gasification processes to facilitate the utilization of coals from different seams or mines while maintaining steady blend quality parameters, e.g.

ash content, sulphur content or heating value. Furthermore, low reactivity petroleum coke, solid residues from chemical processes or low volatile matter (VM) coals are often blended with high VM coal to improve combustion or gasification performance (Shen et al., 2011). Additionally, past and future fossil CO<sub>2</sub> reduction policy as well as high biomass hydrogen content and local availability of waste biomass also encourage an increased thermal utilization of coal-biomass blends.

In the present paper, a comprehensive theoretical study on the potential of binary and ternary fuel blend gasification is carried out, investigating and explaining the gasification reaction kinetic based synergy effect and its impact on key gasification performance indicators. A one-dimensional numerical entrained-flow gasification model is utilized, which is able to process blends up to three different solid fuels. Following the approach of Gräbner & Meyer (2012), the coal blend simulation results are visualized using ternary graphs, to provide a thorough understanding of the synergetic fuel blend effect in gasification. The 1D simulation results will be incorporated in the subsequent 2D-CFD numerical experiments and the detail engineering of the first stage of a novel two stage entrained-dense-plug-flow gasifier design for the processing of fuel blends.

### MODEL DESCRIPTION

The developed model represents a one-dimensional plug flow gasification reactor, hence radial temperature and gas composition gradients are neglected and intra-particle temperature gradients are disregarded. Axial temperature and gas composition profiles emerge consecutively, not allowing for turbulence induced back-mixing of either solid particles, which are usually smaller 300 µm, or gas phase. Specific model development focus is drawn to:

- processing of fuel blends consisting of three different feedstocks (ternary blends);
- high degree of calculation convergence robustness for later sensitivity analysis;
- convergence time limitation to a maximum of four minutes.

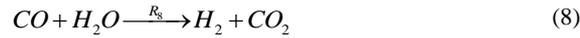
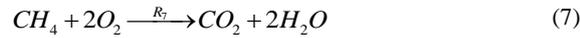
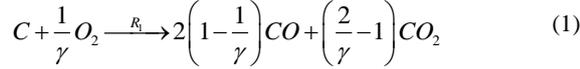
The following sub-models are integrated in the gasifier model:

- drying and devolatilization (product yield and volatile combustion);
- heterogeneous char combustion reaction and heterogeneous char gasification reactions (rate and product yield);
- homogeneous gas phase reactions (rate and product yield);
- char structure development.

Detailed sub-model descriptions can be found elsewhere (Beath, 1996; Liu et al., 2000; Monaghan & Ghoniem, 2012). In the following sub-sections a documentation of the model structure and its implementation within the modelling environment Aspen Plus (AP) is presented. AP has been selected as gasifier modelling environment because of two reasons. (1) The gasifier model can be utilized as part of broader gasification-based process models. (2) AP comprises an extensive physical and thermodynamic properties database.

### Heterogeneous and homogeneous reactions

The following heterogeneous and homogeneous chemical reactions are considered in the gasifier model.



### Conservation equations

Mass balance equations for solid-phase components and gas-phase species are listed in the differential equations Eq. (10) and Eq. (11) below. They represent the rate of mass change along the length of the gasification reactor.

$$\frac{\partial \dot{m}_s}{\partial x} = - \sum_{m=1}^4 \sum_{\beta=1}^3 R_{m,\beta}^{R_1-R_4} M_s \quad (10)$$

$$\frac{\partial \dot{m}_i}{\partial x} = \sum_{m=1}^4 \sum_{\beta=1}^3 (v_i'' - v_i') R_{m,\beta}^{R_1-R_4} M_i + \sum_{m=5}^9 (v_i'' - v_i') R_m^{R_5-R_9} M_i \quad (11)$$

The calculated differential enthalpy change along the reactor length is calculated according to Eq. (12).

$$\frac{\partial \dot{H}}{\partial x} = \sum_{m=1}^9 \sum_{\beta=1}^3 R_{m,\beta}^{R_1-R_9} \Delta h_{m,\beta} \quad (12)$$

### Reaction rates

The heterogeneous reaction rates are calculated based on Arrhenius-type intrinsic reaction kinetic data from Hla et al. (2007) by utilization of an effectiveness factor based approach as shown in Eq. (13).

$$R_{m,\beta}^{R_1-R_4} = \eta_{m,\beta} a_{p,\beta} P_{S,m}^{n_m} k_{m,\beta}^{R_1-R_4} \quad (13)$$

The homogeneous reaction rates are calculated based on Arrhenius-type reaction kinetic data from Varma et al. (1986), Dryer & Glassman (1973) and Silaen & Wang (2010), and are calculated as shown in Eq. (14).

$$R_m^{R_5-R_9} = k_m^{R_5-R_9} \prod_{i=1}^N C_i^{n_{m,i}} \quad (14)$$

### Model structure

The gasification process is divided in major sub-processes (1) drying, devolatilization and char decomposition, (2) volatile combustion and (3) heterogeneous and homogeneous gasification. Fig. 1 illustrates the model structure in AP in a schematic diagram.

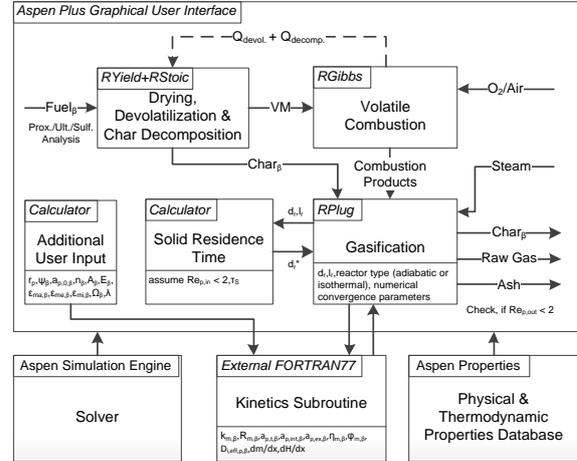


Figure 1: Structure of AP gasifier model

Due to the complex mathematical formulation of the heterogeneous gasification reaction rates including diffusion phenomena, it is necessary to utilize an external FORTRAN77 kinetics subroutine for the calculation instead of using the integrated reaction kinetics calculation form of the RPLUG reactor model in AP. Furthermore, following the approach of Chang (2009), the difference of solid residence time and gas residence time is accounted for by implementation of a calculator block.

### MODEL VERIFICATION

The numerical model is verified with experimental gasification data of 3 Australian coals (CRC252, CRC274, CRC299) from literature (Hla et al., 2007). The experimental setup consists of an air blown, externally heated, pressurized entrained-flow gasification reactor (PEFR). Steady state process conditions are adjusted to coal feed rates usually below 5 kg/h and high nitrogen dilution up to 50 kg/h. Model predictions of carbon conversion in Fig. 2 are in good agreement with the literature data while the gas species development profiles of Fig. 3 - Fig. 5 represent the general formation trends.

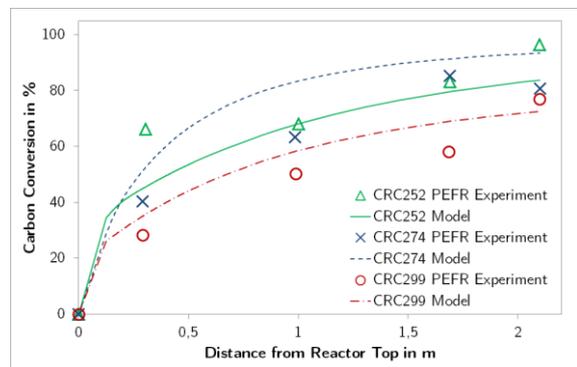


Figure 2: Model carbon conversion and exp. data points

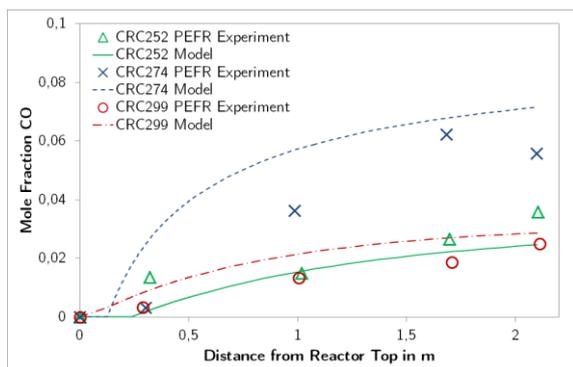


Figure 3: Exp. and model CO mole fraction profiles

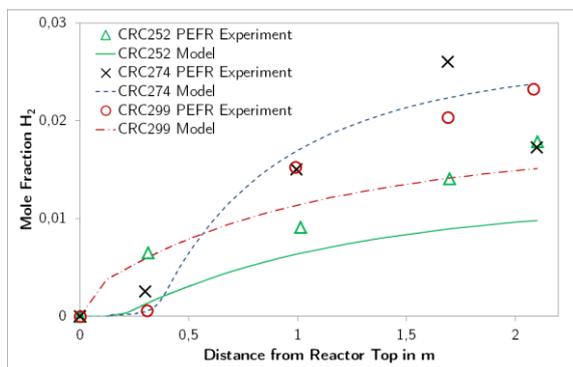


Figure 4: Exp. and model H<sub>2</sub> mole fraction profiles

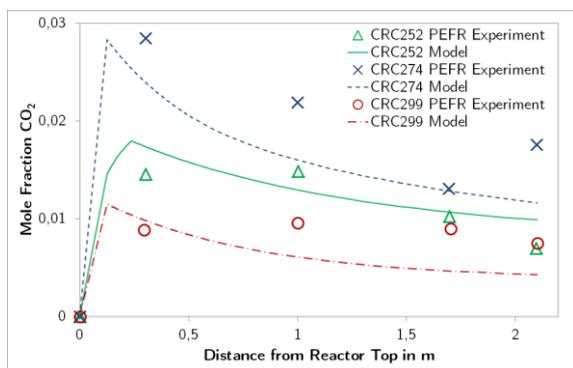


Figure 5: Exp. and model CO<sub>2</sub> mole fraction profiles

The sensitivity analysis of oxygen to carbon molar ratio presented in Fig. 6 demonstrates applicability of the model in a wider range of process conditions.

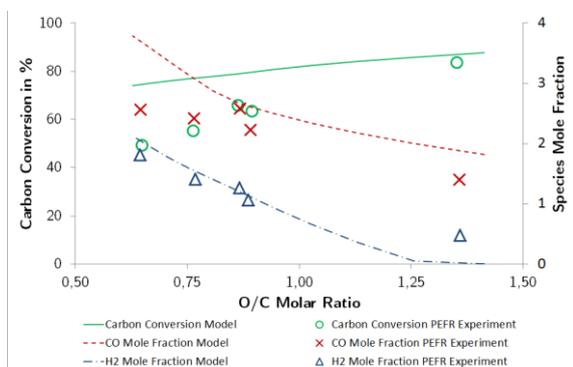


Figure 6: Sensitivity analysis oxygen to carbon molar ratio of model and exp. (CRC274)

## RESULTS AND DISCUSSION

The synergetic fuel blend effect in gasification was experimentally detected when high VM fuels and low VM fuels with high heating value were blended and gasified together. Therefore, the utilized fuel blend in the simulation study consists of a VM rich coal as received (CRC252), char of a low ash coal with high heating value (CRC274 char and char of a high ash coal with comparably low heating value. Detailed fuel analysis data is listed in Tab. 1.

Table 1: Fuel blend component analysis

	Unit	Fuel 1	Fuel 2	Fuel 3
Fuel type		CRC252 (as received)	CRC274 (char)	CRC299 (char)
HHV (Dulong)	MJ/kg (wf)	28.62	29.22	20.82
Proximate analysis				
Fixed carbon	wt. %	39.1	86.2	61.4
Volatile matter	wt. %	38.8	0.0	0.0
Ash	wt. %	11.3	13.8	38.6
Moisture	wt. %	10.7	0.0	0.0
Ultimate analysis				
C	wt. % (waf)	78.1	100.0	100.0
H	wt. % (waf)	5.9	0.0	0.0
O	wt. % (waf)	14.4	0.0	0.0
N	wt. % (waf)	1.1	0.0	0.0
S	wt. % (waf)	0.5	0.0	0.0

The simulation parameters for all carried out calculations are listed in Tab. 2.

Reactor mode	adiabatic
Reactor length	2.1 m
Reactor width	0.07 m
Thermal capacity (const.)	85 kW
$\lambda$	0.3

As a first result Fig. 7 shows the varying conversion results of a binary fuel blend (CRC252-CRC274 char) over the blend ratio. On the left graph side a zone is formed where no ignition of the fuel can be detected. This is due to the lag of VM that is necessary to ignite the fuel.

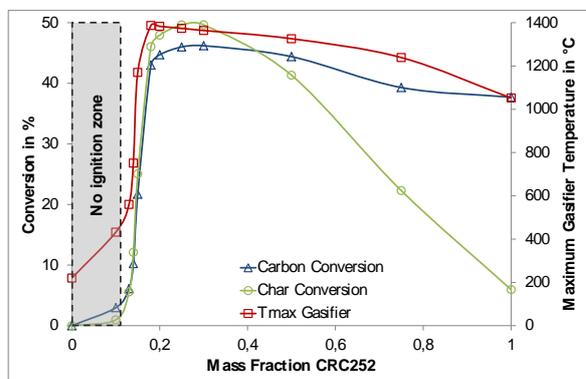


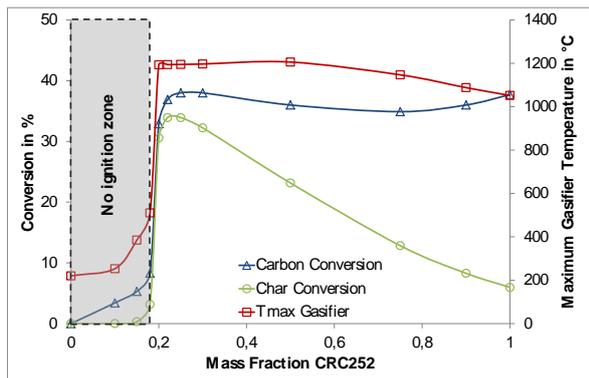
Figure 7: Simulation results of binary fuel blend gasification (CRC252-CRC274 char)

Beyond a critical ratio of 10 wt.% VM rich CRC252, the blend ignites and rapid increase in conversion can be observed with a char and carbon conversion maximum at 30 wt.% CRC252 in the blend. Further increase of CRC252 mass fraction yields a decrease in conversion. This can be explained by the decreasing maximum gasifier temperature caused by a decrease of the fuel influence from high heating value CRC274 char.

The large difference of char conversion values and carbon conversion values can be explained by the amount of VM that increases with increasing mass fraction of CRC252 in the blend. More carbon conversion can be achieved by simple devolatilization and volatile combustion before gasification reactions take place. However, the char conversion from heterogeneous gasification reactions is dependent on the gasifier temperature and steadily decreases with decreasing maximum gasifier temperature. Hence, char conversion profiles provide more valuable information for the characterisation of fuel blend gasification phenomena.

Based on the observation of the binary fuel blend gasification it is possible to characterize the kinetic based synergy effect. By increasing the amount of the VM rich blend component, the activation energy barrier for the blend ignition is exceeded and the blend ignites. Subsequently, the high heating value of the char leads to higher maximum gasifier temperatures than for the VM rich component alone, thus overall conversion is higher. This effect can only be observed when detailed reaction kinetics are accounted for in the simulation since equilibrium based approaches are not able to reveal this effect.

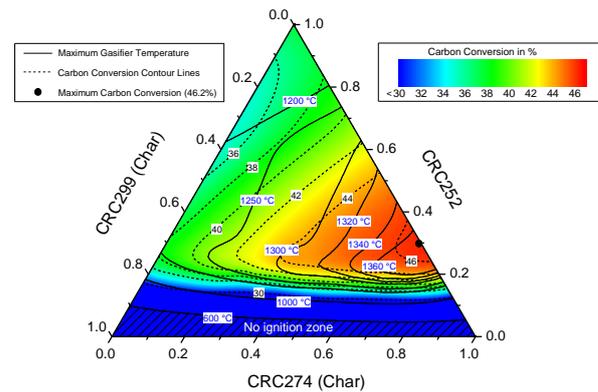
Contrary results can be observed in Fig. 9, where VM rich CRC252 and a high ash, low heating value CRC299 char are gasified as a blend. Here the “no ignition zone” is comparably larger than in the previous graph due to a decreased blend heating value. The conversion maxima are directly connected to the “no ignition zone” and can be found at 20 wt.% CRC252. The highest maximum gasifier temperature is 200 K lower than in the first case because the heating value of the ash rich CRC299-char is low.



**Figure 8:** Simulation results of binary fuel blend gasification (CRC252-CRC299 char)

The binary fuel blend gasification results show that blend ratios for optimal conversion results can be obtained from a simulation analysis. These results can be projected to ternary fuel blend gasification simulations as shown in Fig. 9. A total number of 1000 single simulations were carried out to create this ternary graph which now characterizes a system of the different fuels in a blend. Necessary information about carbon conversion as well as maximum

gasifier temperatures can be predicted and optimal operation conditions can be derived.



**Figure 9:** Simulation results of ternary fuel blend gasification (CRC252, CRC274 char, CRC299 char)

## CONCLUSION

A theoretical study on the potential of binary and ternary fuel blend gasification was carried out and the kinetic based synergy effect was characterized. The obtained information from this study is valuable for the design and operation of gasification plants where different feedstock is utilized and will directly be implemented in the design of a novel 2 stage entrained-dense-plug-flow gasifier.

## NOMENCLATURE

$\dot{m}$	mass flow	(kg/h)
$R$	reaction rate	(kmol/(m s))
$\nu$	stoichiometric coefficient	(-)
$\dot{H}$	total enthalpy flow	(kJ/s)
$h$	reaction enthalpy	(kJ/kmol)
$\eta$	effectiveness factor	(-)
$a$	surface area	(m <sup>2</sup> )
$p$	pressure	(atm)
$k^{R_1-R_4}$	kinetic rate constant	(kg/(m <sup>2</sup> s atm <sup>n</sup> ))
$k^{R_5-R_9}$	kinetic rate constant	(1/s)
$c$	concentration	(kmol/m <sup>3</sup> )

## Subscripts

$s$	solid
$i$	species index
$m$	reaction index
$\beta$	fuel component index
$S$	surface
$p$	particle

## Superscripts

$n$	order of reaction
'	product gas index
"	reactant gas index

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