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# Catalytic effect of potassium in thermochemical gasification of biogenic resources in a floating fixed bed gasifier

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## Abstract

The floating fixed bed wood gasifier has a potential to use low-cost residues from agriculture and forestry. Those feedstocks often contain relevant amounts of potassium salts that are known to lower the ash melting point and to catalytically accelerate the Boudouard reaction, which is one of the main reactions in the gasification process. The influence of varying amounts and types of potassium salts on the reaction rate of the Boudouard reaction was quantified in previous works. Basing on these findings, an ASPEN-Plus®-based tool (SBR-Sim 3.0) was developed to simulate the product gas composition and temperature profile in the floating fixed bed gasifier, caused by potassium-induced changes of the reaction kinetics of the Boudouard reaction.

The main findings from the simulations were that the CO content of the product gas and the carbon conversion rate can be increased when increasing the reaction rate of the Boudouard reaction up to a factor of two, thus improving product gas quality. With a higher enforcement of the reaction rate of the endothermic Boudouard reaction, the reaction lowers the overall temperature in the reactor, thus leading to undesirably increased CO<sub>2</sub> output. Conclusions are drawn on how the potassium content of potassium-containing feedstock will influence product gas quality and temperature profile in the reactor to predict gasification behavior by analyzing the chemical composition of the raw material.

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## 1. Introduction

### Biogenic residues for gasification

The floating fixed bed gasification technology was developed for the energetic use of woodchips in the range of 1 to 5 MW of fuel power. In this staged co-current process, the gravity force of the bulk bed is balanced by the force of the ascending pyrolysis gas flow, which leads to low pressure loss across the bed and avoids the generation of gas channels in the bulk bed. The comparatively long residence time of the pyrolysis gas in the charcoal bed allows an efficient conversion of tars, hence producing

constant gas qualities with tar contents below 50 mg m<sup>-3</sup> (u.s.c) [1].

The use of biogenic residues is of relevance both from an ecological and from an economic point of view. As woodchips represent a high value feedstock that is exposed to high competition regarding its usage (building material, pulp industry), their replacement as a feedstock for energy production by biogenic residues (straw, corn husks etc.) or biogenic wastes (sewage sludge, biogas digestate, waste wood etc.) is desirable. Many of those alternative biogenic raw materials (ABR) are characterized by the following properties:

- The raw material is more inhomogenic than woodchips
- The calorific value is lower than in woodchips
- The ash content is higher than in woodchips
- An increased level of potassium in the ash increases slagging tendencies

These unfavorable properties are contrasted with a lower purchase price, which might increase the economic interest in using ABR. The adaptation of a wood gasification plant to ABR requires considering not only altered fluidic properties caused by varying particle shapes and an altered situation for gas purification but also the slagging properties of raw materials with high potassium content.

The influence of potassium on reaction kinetics, mainly the Boudouard reaction as a representative of heterogenic gasification reaction, was described in previous work [2][3]. The main findings are summarized in this paper. Basing on these findings, an ASPEN Plus® based simulation tool, SBR-Sim 3.0, was developed [4] to simulate the influence of the enhanced reaction rate of the Boudouard reaction on the product gas composition as well as on the temperature profile in the floating fixed bed reactor. [5]

### **Floating fixed bed gasification**

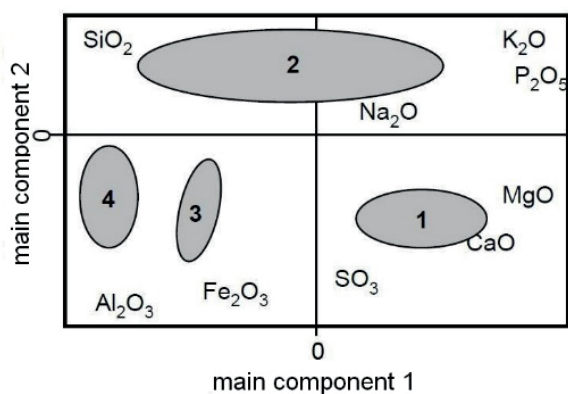
The floating fixed bed gasification is a two-stage autothermal process. In the first stage, the feed material is dried and pyrolyzed at temperatures of 450 to 500 °C. The resulting pyrolysis gas and pyrolysis char are fed to the second stage, the core part of the gasification plant. In this conic reactor, the oxidation and

reduction reactions take place. The floating fixed bed is established by feeding both the gas and the char from the reactor bottom, against gravity. Below the lower edge of the bed, the gasification air is introduced, creating a highly turbulent and partly oxidative zone that provides the required heat for the process. The widening of the reactor towards the top helps to keep the bed floating, saves the need for a grate and avoids sintering of constructive elements. The char particles are thermo-chemically reduced while moving upward in the bed. The product gas is collected at the reactor top and subject to downstream treatment. The basic product gas composition is 17-19 % H<sub>2</sub>, 14-16 % CO, 14-16 % CO<sub>2</sub>, 1,0-2,5 % CH<sub>4</sub>, 47-49 % N<sub>2</sub> (result of > 100 test runs; %vol). Total BTEX range < 100 ppm, total PAH < 10 ppm and total gravimetric tars < 100 ppm.

The constructive design of the reactor allows for low pressure loss (<100 mbar), even material distribution across the cross-section, relatively long gas residence times (5-6 s), and thus avoids local hot spots, material compaction, gas channeling, wall flow and resulting tar creation.

### **Ash melting behavior**

According to Kahn et al. [6], biomass can be classified in six categories following biomass composition, its combustion pattern and ash composition. [7] defines four categories of fuels, based on the oxides of their main ash components, see figure 1. While calcium and magnesium dominate in woody ashes (1), waste materials (3) contain relevant amounts of ferrous oxides. However, it has to be noted that in biogenic materials, in general the component composition distribution may vary substantially.



**Figure 1: Fuel categories, according to their main ash components (1 = woody biomass, 2 = culmiferous (straw-based) biomass, 3 = waste materials, 4 = chars) [7]**

Ash melting patterns are characterized by four standardized specific temperatures (shrinking starting temperature, initial deforming temperature, hemisphere temperature, flow temperature). The complex reaction schemes of biogenic feedstock ash components are difficult to predict. Several authors, such as [7][8][9], made efforts to conclude the ash melting pattern from the ash composition. However, most findings are basing on ashes from combustion plants. With gasification, also a high carbon content can be found in the ash. The remaining carbon influences the ash melting [5], and due to the diversity of biomass compositions, a defined ash melting point does not exist.

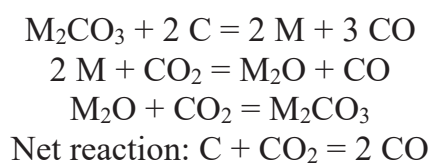
### Potassium as a catalyst

Potassium has a history of use as a catalyst in coal gasification for acceleration of heterogenic reactions [10][11]. Numerous publications confirm the catalytic properties of potassium in coal gasification reactions. It catalyzes the heterogeneous water-gas reaction of activated carbon in the same way as a metal oxide catalyst [10]. It improves the oxygen transfer from the gas phase to the carbon surface, thus increasing the number of adsorptive bonds of carbon to oxygen and to carbon monoxide. Hence, the

carbon-carbon bonds are weakened and reactivity is enhanced [12]. Potassium and sodium carbonate catalyze the Boudouard reaction mainly at temperatures below 1000 °C, while above 1000 °C non-catalytic mechanisms are effective [13]. The reaction rate of the heterogenous water-gas reaction is enhanced by a factor of 10 when adding 10 %m potassium carbonate to a char gasification with steam and CO<sub>2</sub>. At a constant carbon conversion rate, the gasification temperature could be lowered below 700 °C [14].

At steam gasification of spruce, a 3.0 mmol alkali supplement of potassium carbonate per g spruce increased carbon turnover from 77 to 93 % [15]. At examinations of potassium in various biomass ashes it was found that the catalytic activity of potassium is decreased by silicon. It is assumed that potassium silicates are formed, which deactivate the catalytic mechanisms [16].

At the Boudouard reaction, the catalytic effect of potassium is described by two mechanisms. One of those is relevant for this paper and restricted to temperatures below 800 °C. The reaction steps are described as follows with M representing any alkali metal [17]:



The mechanism shows that the reaction steps could run infinitely without the alkali carbonate being depleted [18].

In the context of gasification of low melting raw materials it was investigated if and to what extent the gasification temperature in the reactor can be lowered in order to achieve a carbon conversion rate that is equivalent to wood gasification,

considering the temperature-dependent longer residence time in the floating fixed bed reactor [5]. Therefore, various biogenic raw materials with varying potassium contents as well as potassium-added spruce wood samples were gasified in the lab under CO<sub>2</sub> and steam atmospheres, respectively, to determine the carbon turnover reaction velocity compared to raw spruce wood. The data base for the ASPEN Plus®-based simulation tool SBR-Sim 3.0 was fed with the results of those examinations. Carbon turnover and relative reaction velocity were investigated by thermogravimetric analysis (TGA) [3]. Pyrolyzed pellets of wood (partly supplemented by various potassium salts) and biogenic residues were gasified under CO<sub>2</sub> atmosphere. It was found that by adding up to 5 %m of potassium, at 850 °C, the reaction velocity multiplies up to the quadruple. The catalytic effect of potassium could not be quantified exactly because of strong dependency on the type of potassium salt used. However, it is concluded that the raw material's composition already suggests its reactivity [2][3]. As the possibility exists that potassium silicates (eutectica) are formed that may further lower the melting point [16][19] and as well back-influence the catalytic effect of potassium, the effect of silicon content in samples is highlighted in [3].

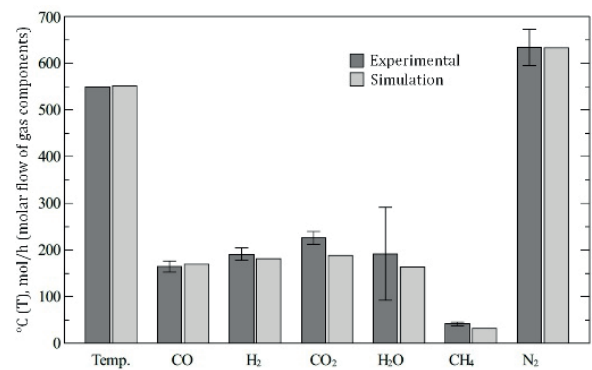
## 2. Concept and methodology

The standard model of ASPEN Plus® does not cover kinetics and residence time data. The calculations result from thermodynamic equilibria. The heterogeneous water-gas reaction is a rather slow reaction, and it can be assumed that the residence time in the respective temperature zone in the reactor is not long enough to achieve equilibrium. Thus, kinetics had to be programmed additionally by Fortran modules. Hence,

the following reactions were completed by kinetic data [4]:

- Carbon oxidation
- Hydrogen oxidation
- Boudouard reaction
- Heterogeneous water-gas reaction
- Hydrogenating gasification reaction
- Water-gas shift reaction

Starting from the version SBR-Sim 2.x that still produced strong deviances in the gas composition, the model was refined by kinetic data from literature and the reactivity of pyrolysis char via the CRF (char reactivity factor) [20]. The updated SBR-Sim 3.0 shows only small deviations from real experimental data of the pilot gasification plant with a fuel power of 250 kW. The comparative results are shown in figure 2.



**Figure 2: Product gas temperature and composition: Experimental (VL CW Alpha) and Simulation (SBR-Sim 3.0)**

The simulation design covers the particularities of operation and reactor type. Phase ratios, temperature control and residence time patterns cannot be reproduced through one of the standard appliances provided by the simulation software. Due to the strong variations of temperature and concentrations across the reactor height, the floating fixed bed reactor may best be compared to a plug flow reactor.



In addition to temperature and concentration changes of all reactants, also particle sizes, bulk and particle porosities as well as particle surface areas change as particles move through the reactor. Due to the conic reactor geometry, also the gas-particle residence time and the inflow velocity vary across the reactor height. This is why a cascade of stirred tank reactors is chosen for the simulation model. It is assumed to reflect the prevalent reaction conditions in relation to the respective site in the reactor. It also allows to assign higher definition to more relevant zones of the reactor such as the inlet or the lower edge of the charcoal bed, and to simplify less relevant parts. 14 continuously stirred tank reactors (RED-1 to RED-14) were defined, starting at the reactor bottom where the feed and gas inlets are located (see figure 3).

In the simulation model, the following assumptions were made:

- The simulation covers only the main floating fixed bed reactor, i.e. oxidation and reduction processes. Upstream drying and pyrolysis and downstream dust filtering, gas washing etc. are neglected.
- The feed stream is composed of pyrolysis gas and char and based on empirical data.
- Due to its special structure and the inherent several reaction zones, the reactor is modeled as a series of cascading stirred tank reactors.
- Only the steady state is investigated. Radial temperature and concentration gradients are neglected.
- The main reactions are given with the relevant kinetic data. Any further

intermediate and side reactions will be calculated by the simulation software.

- Particles are handled according to the shrinking core model.

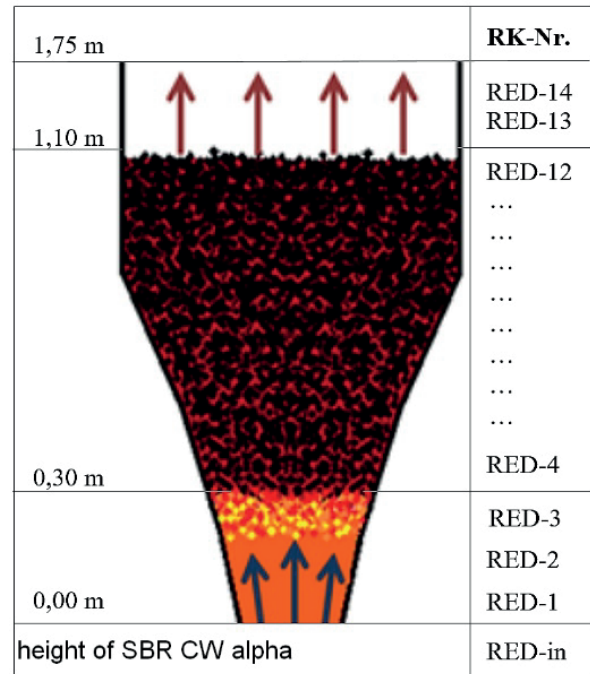


Figure 3: Modeling the reactor segmentation

For the simulation, the Boudouard reaction is considered a pseudo first order reaction (carbon is a solid available in excess). In this case, the reaction velocity  $r_i$  is a product of reaction velocity constant  $k_i$  and the concentration of carbon dioxide, and hence a factor to increase the reaction velocity equates to a factor of  $k_i$  (see eq. 1-3).

$$r_{C-CO_2} = k_{C-CO_2} \cdot c_C \cdot (p_{CO_2} - p_{CO_2}^*) \cdot V_{ST} \quad (1)$$

$$k_{C-CO_2} = k_{0, C-CO_2} \cdot e^{-\frac{E_{A, C-CO_2}}{R \cdot T}} \quad (2)$$

$$p_{CO_2}^* = \frac{p_{CO_2}}{e^{\frac{20,92}{20280} + \frac{20280}{T}}} \quad (3)$$

with

- $r_{C-CO_2}$  reaction rate in  $\frac{\text{mol}}{\text{s}}$
- $k_{C-CO_2}$  reaction velocity constant in  $\frac{1}{\text{atm} \cdot \text{s}}$
- $c_C$  molar carbon concentration in  $\frac{\text{mol}}{\text{m}^3}$
- $p_{CO_2}$  partial pressure of carbon dioxide in atm

$V_{ST}$  stirred tank volume in  $m^3$   
 $k_{0, C-CO_2} = 930 \frac{l}{atm \cdot s}$   
 $E_A = 45.000 \frac{cal}{mol}$   
 $R = 8,314 \frac{J}{mol \cdot K}$   
 $T$  temperature in K

A schematic description of the routine of the simulation model can be retrieved from figure 4.

At constant temperature,  $k_i$  depends of the frequency factor  $k_0$  and the activation energy  $E_A$ . By setting that parameter in ASPEN Plus® via Fortran code, the underlying kinetics can be influenced. It is assumed that the catalytic activity of potassium does not influence the steric factor significantly but intervenes only in activation energy.

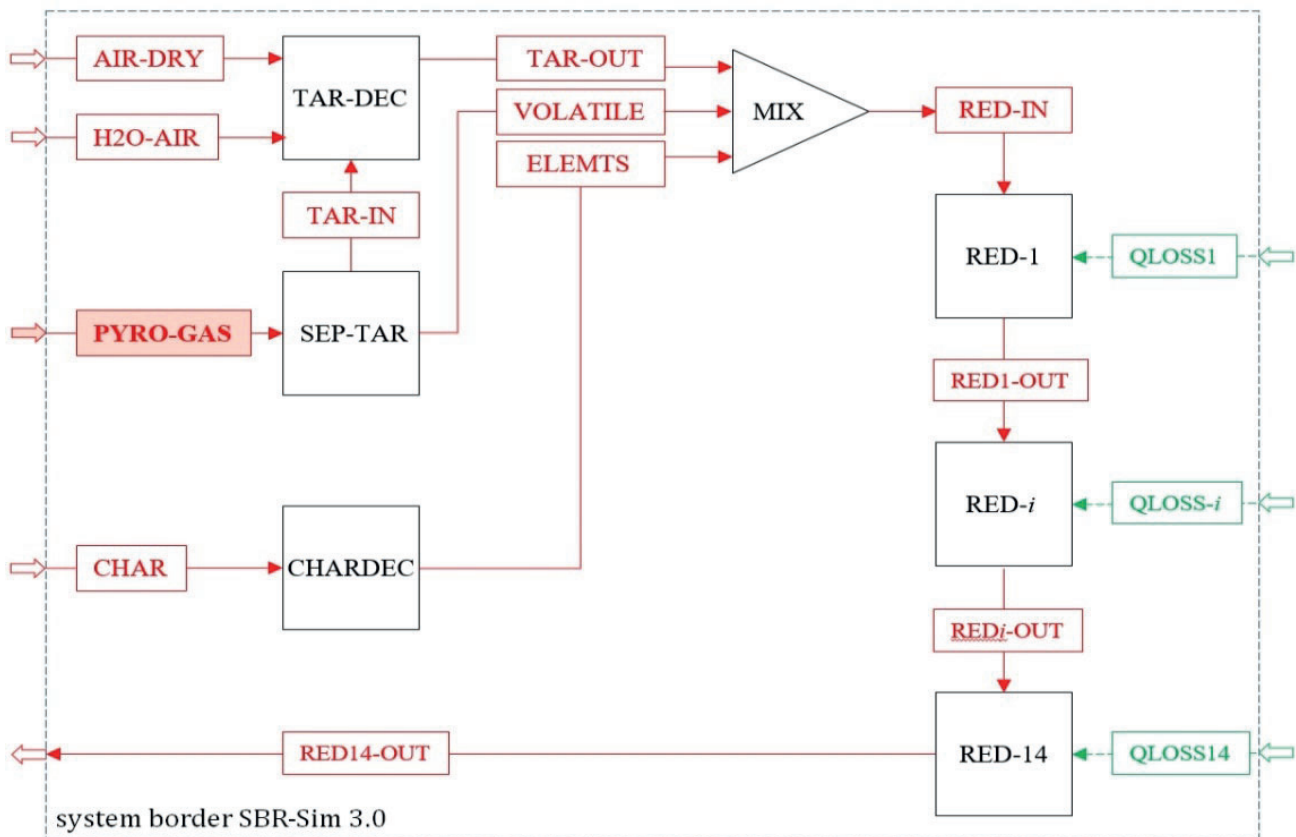
Thus, the respective activation energy can be calculated. The parameters of the simulation runs are listed in table 1. For the intervention in the underlying kinetics,

the Fortran code has to be adapted and compiled before each simulation run.

**Table 1: Kinetic parameter of simulation runs K 1.1 to K 1.6**

	Factor of $k_i$	$E_A$ in $\frac{cal}{mol}$
K 1.1	1,00	45.000
K 1.2	1,25	42.955
K 1.3	1,50	41.285
K 1.4	2,00	38.648
K 1.5	3,00	36.606
K 1.6	5,00	34.932

Material streams are marked red, calculation modules black and energy loss streams green. The main input stream is split to PYRO-GAS and CHAR streams. The PYRO-GAS stream is defined by real pilot test data. Tars contained in pyrolysis gas are referred to as benzene.



**Figure 4: Process, modelled in SBR-Sim 3.0**

According to pilot test results, tar cracking is an upstream process of the TAR-IN (benzene) stream, effected in TAR-DEC at air/750°C. The resulting product stream is TAR-OUT. The solids stream CHAR is defined a non-conventional solids stream that does not participate in chemical equilibrium. CHAR is decomposed into its components at CHARDEC. The resulting ELEMETS stream contains a conventional elemental stream of reaction partners.

The gasification air consists of a mixture of AIR-DRY and H2O-AIR.

The streams resulting from those upstream processes are mixed (MIX) and fed to the reactor as RED-IN. The reactor is segmented into a cascade of 14 separately parametrized continuous stirred tank reactors (CSTR, RED-*i*) that the streams pass and leave as RED14-OUT. Heat losses are referred to as negative input streams QLOSS-*i*.

Based on the simulation of the VL CW Alpha [20] and the kinetics proposed in [21] for the Boudouard reaction, the reaction velocity is varied by the factor of 1 to 5.

The profiles of temperature, gas composition and calorific value of the product gas are displayed in relation to the height of the reactor. The achievable carbon conversion rate and cold gas efficiency of the respective simulation runs are compared. The cold gas efficiency rate  $\eta$  signifies the heating value  $H_i$  of the product gas mass flow  $\dot{m}$  in relation to the energy content of the biomass per time unit (eq. 4):

$$\eta_{CG} = \frac{H_{i, \text{product gas}} \cdot \dot{m}_{\text{product gas}}}{H_{i, \text{biomass}} \cdot \dot{m}_{\text{biomass}}} \quad (4)$$

All experimental raw data can be retrieved from the authors on request.

### 3. Results and discussion

The following figures 5 and 6 show the characteristics of material flow temperature and mole flows of product gas components over the reactor height of the respective simulation runs at standard reaction velocity (factor = 1, figure 5) and 5 times increased reaction velocity (factor = 5, figure 6). The figures show that both the gas composition and the temperature at the outlet of the reactor are more or less in the same range in both cases. The big difference can be seen in the concentration and the temperature profile over the height of the reactor. With the standard reaction rate, the resulting maximum temperature in the lower zone of the reactor is at 870 °C. When rising the reaction rate by factor 5, the maximum achievable temperature is lower than 700 °C due to the enhanced endothermic reaction, and the temperature profile is more homogeneous with the standard reaction velocity. In contrast, the gas composition, mainly the mole fractions of CO and CO<sub>2</sub>, are more homogeneous over the height in case of the higher reaction rate of the Boudouard reaction.

Figure 7 shows the temperature and dry gas heating value graphs over the reactor height of all simulation runs. The desired maximum temperature of 800 °C to avoid slagging in the reactor is indicated as a vertical line.

The simulation results demonstrate that an increase in Boudouard reaction velocity at first lowers the temperature in the whole reactor, as expected. When increasing the reaction velocity by a factor of 2, the critical temperature of 800 °C is not exceeded at any site in the reactor.

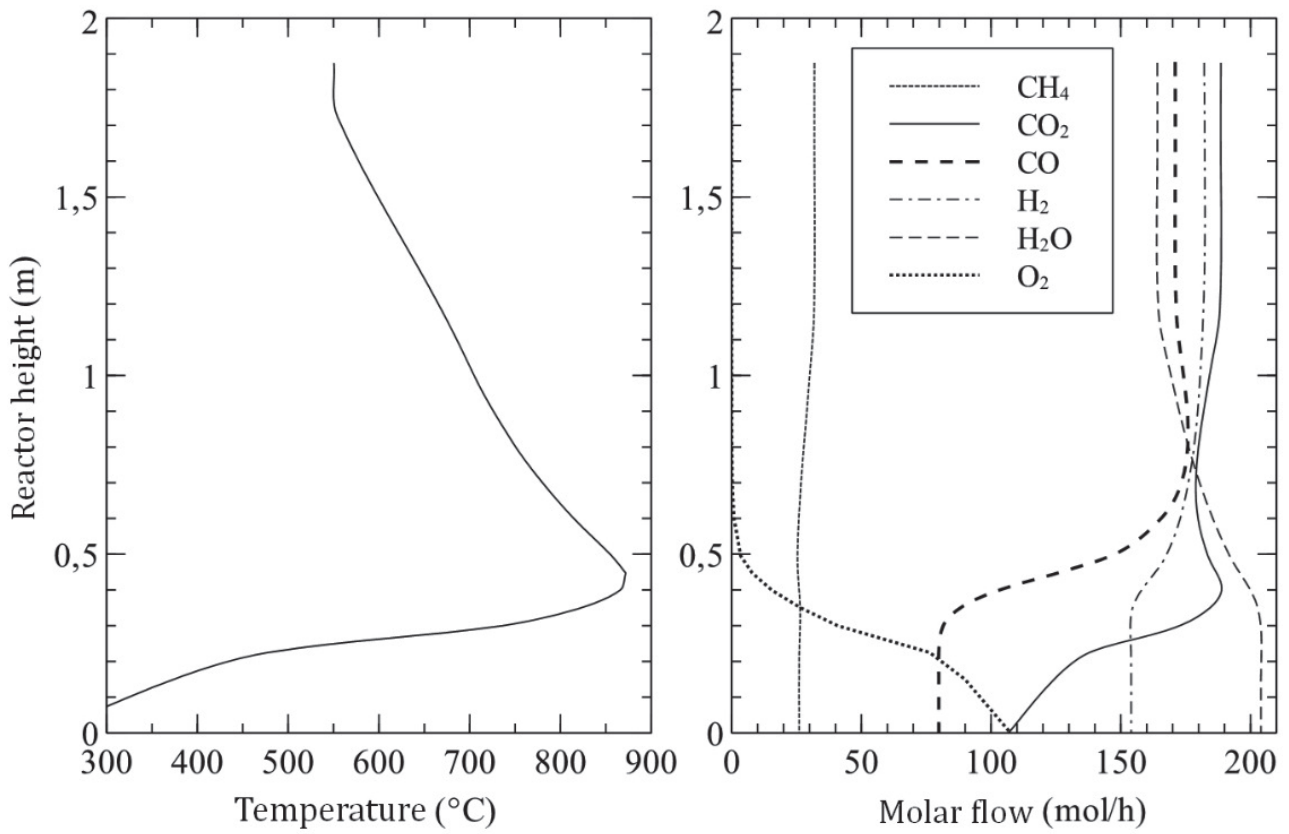


Figure 5: Temperature,  $\dot{n}_{gas}$  at respective reactor height - K 1.1, factor = 1

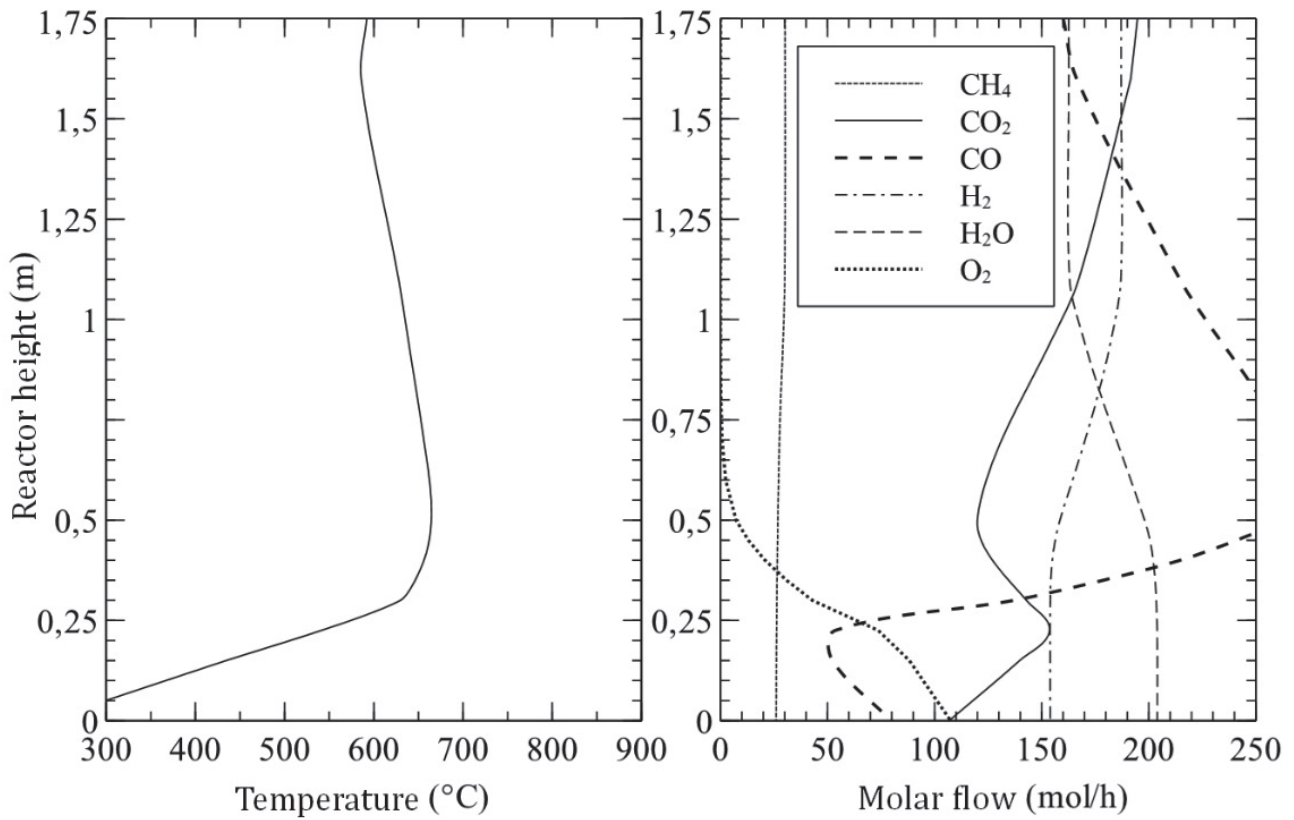


Figure 6: Temperature,  $\dot{n}_{gas}$  at respective reactor height - K 1.6, factor = 5



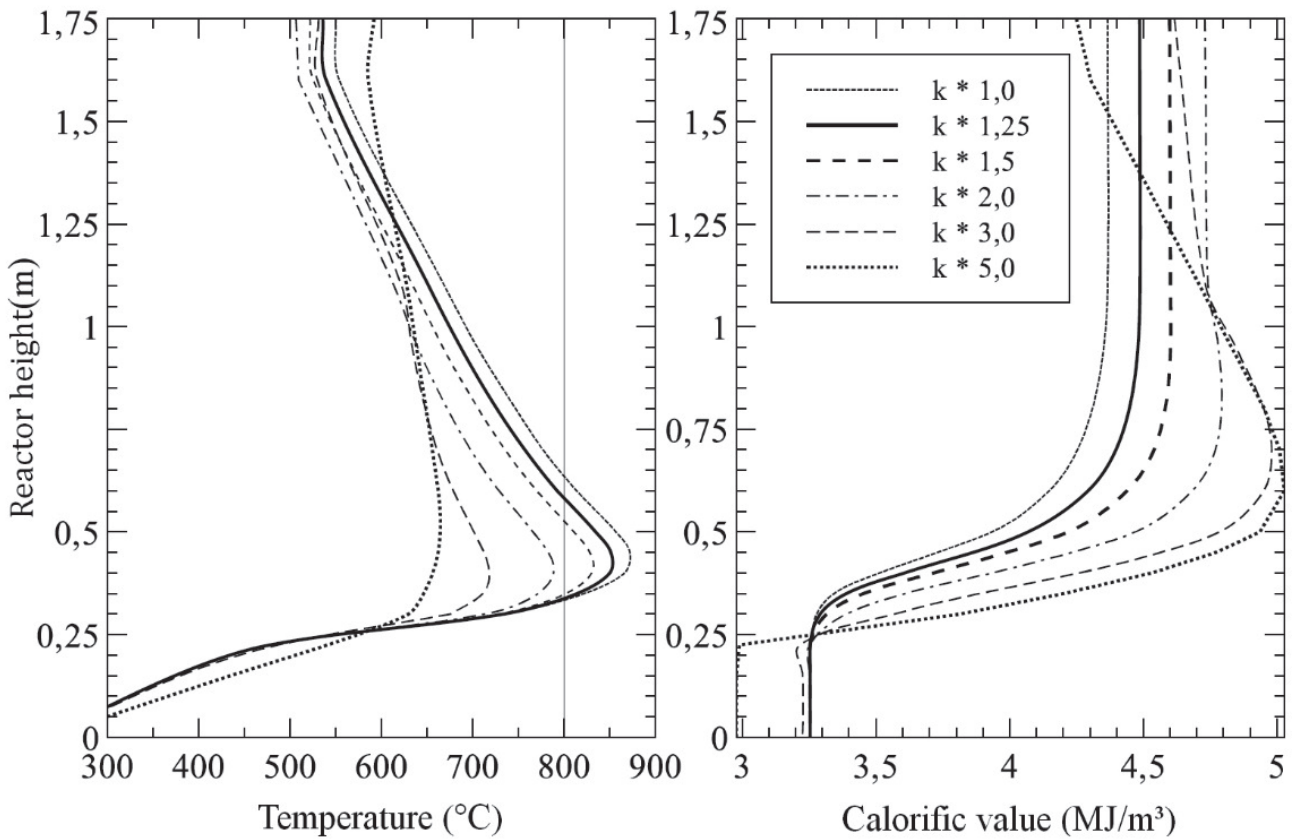


Figure 7: Temperature and heating value as a function of reaction velocity

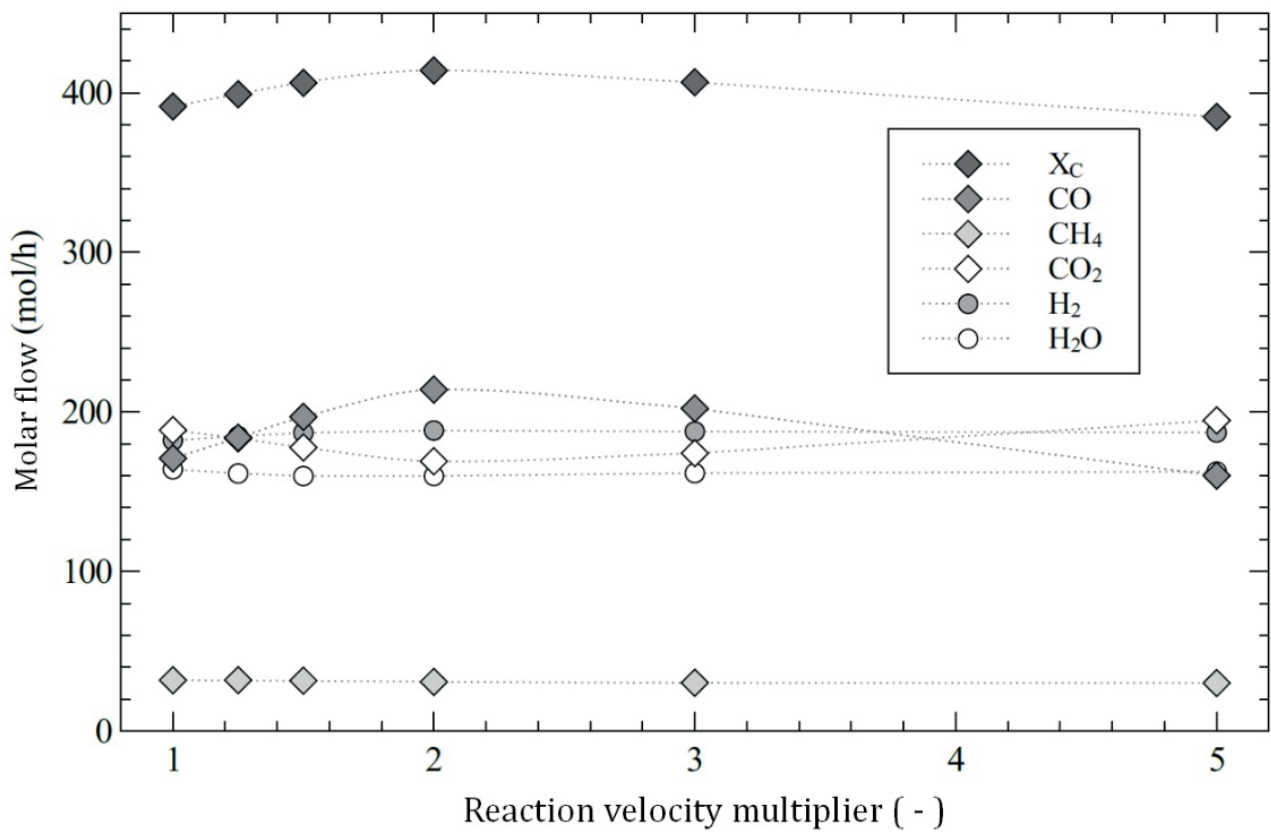


Figure 8: carbon conversion rate  $X_C$  and mole flows of product gas components

With increasing reaction velocities, the temperature profile changes: In the lower reactor segments RED-1 to RED-10 the temperature continues to drop with the factor of the reaction speed, while in higher segments (real reactor height > 0,8 m), the temperature is rising with a factor of 5. This might be due to a shift of Boudouard reaction equilibrium at the point where the originally endothermal reaction to carbon monoxide is inverted towards an exothermal reaction to carbon dioxide. The phenomenon can be deduced from the product gas composition, as the carbon monoxide content declines strongly while the CO<sub>2</sub> content increases accordingly.

Figure 8 shows the product gas composition at the reactor exit as well as the carbon conversion rate  $X_C$  in mol h<sup>-1</sup> as a function of reaction velocities.

The maximum of CO and the minimum of CO<sub>2</sub>, respectively, can be found at the velocity factor of 2. This also leads to a maximum carbon conversion rate at the same point.

The results confirm that the potassium-induced increase of reaction velocity of the Boudouard reaction influences the carbon conversion, the reaction temperature and the product gas composition. Increasing the reaction velocity up to a factor of 2, which corresponds to a potassium content of 2.5 %m in the ash, benefits the gasification process in the chosen simulation model. At higher potassium contents, which lead to higher reaction velocity, endothermal reactions get more relevant and thus, temperatures in the reactor drop. Hence, the CO/CO<sub>2</sub> equilibrium is pushed towards CO<sub>2</sub>, leading to a deterioration of product gas quality.

Although there are also catalytic effects described on the heterogeneous

water gas reaction [14], these influence could not be observed in the setup of the floating fixed bed gasification reactor, neither with experiments on the pilot plant nor in the simulation. A reason therefore could be the lower reaction speed of this reaction in combination with the low gas residence time at temperatures higher than 750 °C [4][20].

#### 4. Conclusion and Outlook

Residues from agriculture and forestry have a high potential to be used as secondary raw materials in the energy industry. Due to their high potassium content and resulting low ash melting point, the utilization in thermo-chemical processes is restricted by technical viability. Economic advantages resulting from low procurement prices are diminished by higher cost for pretreatment and for technical adaptations of the processing plant.

The thermochemical gasification in the floating fixed bed reactor offers a certain potential to benefit from the catalytic properties of potassium containing ABR by lowering the maximum temperature in the reactor and thus avoiding the slagging temperature range. The gasification simulation by means of the SBR-Sim 3.0 simulation tool confirmed the literature data regarding the effects of potassium-enhanced reaction kinetics of the Boudouard reaction. Thus, a raw material's chemical ash composition allows to estimate if and to what extent the Boudouard reaction will be accelerated. However, an exact prediction is not possible due to the complexity of ash transformation reactions. Any potential raw material has to be analyzed by thermogravimetric analyses for a reliable statement on the extent of catalytic effects.

It has to be considered that at lower reaction temperatures, the tar reduction by

thermal cracking across the bulk material is influenced. The residence time is slightly reduced in segments above 700 °C. For the moment, it is impossible to quantify the fraction of thermal cracking on total tar reduction in the floating fixed bed reactor. It is assumed that the main fraction of tars is directly oxidized at the entrance site of the gasification air, and that the final elimination takes place across the bulk material [22]. Thus, it is possible that the remaining tar content is still much lower than at other gasification processes, even if the process is running at low temperature. However, a rise in tar content compared to standard operation mode has to be expected.

The results show that the increased reaction velocity of the Boudouard reaction to some extent positively affects the product gas quality in the floating fixed bed gasification process. The simulation tool will support further process improvements. For the future development, a model validation using real test data is indispensable. The simulation results can be considered as plausible, and they can be understood as a possible trend, as it was not yet possible to

match the simulation to real test operation results. For validation purposes, the pilot plant CW Alpha has to be adapted to meet the simulation parameters.

The low temperature gasification of potassium containing raw materials with the floating fixed bed gasification process is technically viable. However, losses in product gas quality and cold gas power have to be expected. The final decision will be an economic decision that has to be taken specifically for each raw material.

### Abbreviations

ABR	Alternative biogenic raw materials
BTEX	benzene, toluene, ethylbenzene, xylene
CRF	char reactivity factor
CSTR	continuous stirred tank reactor
CW	CraftWERK, trade name
PAH	polycondensed aromatic carbohydrates
TGA	thermogravimetric analysis
u.s.c.	under standard conditions
VL	test run (Versuchslauf)
%m	mass fraction
%vol	volume fraction

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