Co-gasification of Plastics and Biomass in a Dual Fluidized-Bed Steam Gasifier: Possible Interactions of Fuels

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ABSTRACT: Co-gasification of biomass and plastics was investigated in a 100 kW dual fluidized-bed pilot plant using four types of plastic material of different origins and soft wood pellets. The proportion of plastics was varied within a broad range to assess the interaction of the materials. The product gas composition was considerably influenced by co-gasification, whereas the changes were nonlinear. More CO and CO2 were measured in the product gas from co-gasification than would be expected from linear interpolation of mono-gasification of the materials. Less CH4 and C2H4 were formed, and the tar content in the product gas was considerably lower than presumed. With the generation of more product gas than expected, co-gasification of wood and plastic materials also had other beneficial effects. Because of the fuel mixtures, more radicals of different types were available that interacted with each other and with the fluidization steam, enhancing the reforming reactions. Wood char had a positive effect on polymer decomposition, steam reforming, and tar reduction. As a result of the more active splash zone during co-gasification of wood and plastics, contact between gas and bed material was enhanced, which is crucial for catalytic tar removal.

INTRODUCTION

An innovative and viable technology for the thermal conversion of biomass, gasification has been the focus of considerable research for a number of decades. Different gasification systems have been investigated, developed, and successfully implemented in the market, so that several types of gasifiers can be considered a proven technology by now. For the economic operation of a gasification plant, feedstock costs play a decisive role. Waste materials as alternative fuels are therefore receiving increased attention, also for the operation of biomass gasification plants. Plastic wastes are an interesting option, because they are available in large volumes. Mainly originating from the packaging of consumer goods, plastics currently represent 11% of municipal solid waste (MSW) in OECD countries. Plastic wastes are most commonly disposed of together with the mass of municipal waste, although 33% of plastic packaging was already being recycled in the European Union (27 member states) in 2010. In the scope of sustainable resource use, the recovery and/or reuse of waste materials is the most desirable option in place of landfill or incineration without further production of heat and power. For plastic materials, the reuse of carbon or larger molecules via chemical recycling is especially promising. A gasification process involving an extended range of feedstock that can use biomass as well as plastics would therefore offer an innovative approach to the chemical recycling of plastics. In addition to economic benefits for the operators, co-gasification could also bring about synergistic effects because of the use of feedstock mixtures.

Co-gasification of biomass and plastics has been studied in reactors of different types and scales. To describe the interaction of the two materials, all phases of thermal conversion have to be considered, including devolatilization, gasification of carbon, and secondary gas–gas reactions. Devolatilization without secondary reactions can be studied using thermogravimetric analysis (TGA) devices, whereas pyrolysis experiments can be employed to illustrate the combined effect of devolatilization and secondary reactions of volatile matter. Gasification tests involve analysis of both the gasification of solid carbon and the influence of the gasification agent. Therefore, a short overview on the literature is given on all of these important aspects for gasification.

Several TGA studies using biomass and plastics have been published. Jakab et al. investigated the effect of wood, cellulose, lignin, and activated charcoal on the thermal decomposition of polystyrene (PS) and polyethylene (PE) using thermogravimetry with mass spectroscopy (TG–MS). The differential thermogravimetry (DTG) curves for wood, lignin, and cellulose show that these materials decompose at lower temperatures than PS and PE. The charring reactions of the lignocellulosic materials continue in the temperature range of PS and PE degradation. Although the DTG profiles of the lignocellulotics are not affected by the presence of PS or PE, the DTG maximum of PS shifts to higher temperatures in the presence of all wood-derived additives. In a similar study, Jakab et al. examined the thermal decomposition of polypropylene (PP) in the presence of wood flour, lignin, cellulose, and charcoal. PP was mixed with pure activated charcoal to simulate the effect of freshly formed pyrolytic chars. Similar to PS and PE, PP decomposition starts later than decomposition of the other materials in TG–MS. The presence of charcoal accelerates the decomposition of PP. Sharypov et al. tested beech wood, pine wood, cellulose, and hydrolytic lignin in mixtures with PE, isotactic PP, and atactic PP. Biomass degrades at lower temperatures than polylefins, but the mutual influence of biomass and plastics is not apparent in TGA. Dong et al. produced similar results for different mixtures of sawdust.
and PE in a TG–DTG system. In this case, the devolatilization of mixtures of pine and PE is found to be treatable as the linear combination of the pure substances.

In all of the aforementioned TGA studies, the decomposition of biomass and plastics occurs sequentially and, therefore, the interaction of the materials is rather minor. Although biomass decomposition is not affected by the presence of plastics, the char from biomass accelerates the decomposition of PP and hinders the decomposition of PS. The behavior of mixtures can nevertheless be approximated using linear combinations of pure substances, with good agreement.

In contrast to TGA studies, flash pyrolysis experiments involve higher heating rates being applied to samples; as a consequence, biomass and plastics decompose simultaneously and, thus, interactions are more probable. Co-pyrolysis of biomass and plastics has been studied in different reactors, including pyrolyzers for GC–MS, autoclaves, and fluidized-bed reactors. Jakab et al.\(^3,4\) used a pyrolyzer coupled to GC–MS to perform flash pyrolysis in a small quartz tube heated to 550 °C. When studying the effect of wood, cellulose, lignin, and activated charcoal on PS and PE,\(^7\) the authors reported changes in the product distribution related to the different amounts of char produced by each of the lignocellulosic materials. Charcoal is found to promote the hydrogenation of unsaturated products. It is assumed that unsaturated products, such as alkadienes, are absorbed onto the surface of the charcoal, where they are hydrogenated and then released at higher temperatures. In the case of pyrolysis of a 1:1 mass mixture of PP and activated charcoal,\(^7\) fewer oligomeric products are generated than during the pyrolysis of pure PP, while the formation of products of lower molecular weight, including monomers and dimers, is promoted. Dong et al.\(^6\) employed a U-shaped tube reactor for flash pyrolysis at 800 °C to test mixtures of pine and PE (0, 33, 66, and 100% pine) and observed strong interactions of volatiles during pyrolysis. CO formation decreases nonlinearly with an increase in PE in the mixtures. More large hydrocarbons are produced. This increase is also nonlinear with the proportion of PE. In their study of slow pyrolysis using a rotating autoclave pressurized at 3.0 MPa and 400 °C, Sharpyov et al.\(^5\) found that co-pyrolysis of beech or cellulose with PP led to high yields of light distillate. Non-additive effects are also observed when mixtures with more than 50% PP are used. The yields are more than 2 times higher than those expected from the pure substances. Paradela et al.\(^8\) also used an autoclave for co-pyrolysis at 0.41 MPa and 400 °C, employing a typical waste plastic mixture of PE, PS, and PP. This plastic mixture was added to pine in different proportions with the aim of improving slow pyrolysis of pine. An increase in plastics in the mixtures is found to result in a nonlinear increase in the liquid yield and an almost linear decrease in solid products. In contrast, the gas yield decreases steeply with an increase in plastics, as less CO and CO\(_2\) are formed. Berrueco et al.\(^8\) used a bubbling fluidized-bed reactor to investigate the co-pyrolysis of PE and pine. Whereas pyrolysis of PE alone generates alkenes, alkanes, and aromatics but no oxygenated compounds, pyrolysis of pine produces acetic acid, ketones, phenols, CO, and CO\(_2\). During co-pyrolysis of a 1:1 mass mixture of pine and PE, more gas is formed than expected from the pure substances. This could be related to the greater importance of the reactions taking place in the freeboard. Smaller amounts of H\(_2\) and CO are formed from PE than from pine, while H\(_2\) and CO increase nonlinearly in the mixture. Reduced tar formation during co-pyrolysis indicates a synergistic effect on the decomposition of the two materials, possibly associated with char particles produced from the biomass.

Although the aforementioned co-pyrolysis studies employed different reactors and both slow and fast pyrolyses were applied, they all provide evidence of the synergistic effects of biomass and plastic materials. Nonlinear phenomena are commonly observed that would be underestimated if only tests with single substances were carried out, adding the contribution from each feedstock according to its weight ratio in the mixture.

During gasification, the devolatilization products and char can interact with each other and with the gasification agent. Several research groups have investigated the co-gasification of biomass and plastics using fixed-bed or bubbling fluidized-bed reactors. Ahmed et al.\(^9\) studied steam gasification of PE and wood chips in a semi-batch fixed-bed reactor at 900 °C. The authors employed four different mixtures of PE and wood chips to evaluate the influence of possible feedstock interactions. An increase in the PE content in the mixture results in the production of more gas, including higher yields of H\(_2\) and C\(_2\)H\(_4\), as well as increased carbon conversion and energy content of the gas. These increases are all nonlinear; linear interpolation underestimates gas, H\(_2\), C\(_2\)H\(_4\), and energy yield. The maximum gas yield is reached using 80% PE.

Pinto et al.\(^10\) studied systematically co-gasification of biomass and plastics in a steam-blown bubbling bed gasifier, using different mixtures of pine and PE (up to 60 wt % PE) as feedstock. With an increasing proportion of PE in the mixture, more H\(_2\) and C\(_2\)H\(_4\) and smaller volumes of CO and CO\(_2\) are produced. The observed change in gas composition is nonlinear and steep for a PE content of up to 20 wt %. At higher proportions of PE, the gas composition remained almost unchanged. The greatest gas yield is reached using 60 wt % PE. Ruoppolo et al.\(^11\) compared the gasification of pine to that of pellets made from a mixture of 20 wt % PE and pine, using a bubbling bed gasifier fluidized with air and steam. More CH\(_4\) and tar are produced from the mixture of PE and pine.

Mastellone et al.\(^12\) and Pinto et al.\(^13\) investigated various mixtures of coal, biomass, and plastics in bubbling fluidized-bed gasifiers with air or air and steam fluidization. These studies provide a broad overview as to potential feedstock mixtures for co-gasification and summarize the influence of many different factors involved in the process. Mastellone et al.\(^12\) attributed an increased gas yield and a lower calorific value (LCV) of product gas and tar formation to the presence of plastics in the feedstock. Pinto et al.\(^13\) found an increase in plastics to result in greater concentrations of CH\(_4\) and tar and reduced H\(_2\) production.

The cited literature provides evidence of the nonlinear effects that may occur during the co-gasification of biomass and plastics. It is important to investigate feedstock mixtures because the nature of the associated synergistic effects cannot be hypothesized from gasification of the pure substances. During gasification, product gas composition and process performance are influenced by many factors, including the gasification temperature, type of gasification agent, type of bed material, and reactor type and design. TGA and pyrolysis studies have shown that the presence of biomass char influences the thermal conversion of polymers. As a consequence, the distribution of fuel, gas, and char in the reactor is also of importance.

This paper presents the results of a comprehensive investigation into the co-gasification of biomass and plastics in a dual fluidized-bed (DFB) steam gasifier. Four different types of plastics were used in the experiments, both waste
materials and virgin polymers: plastics from MSW, plastics from automotive shredder residues, PE regrind, and virgin PE. Different mixtures of plastics and soft wood pellets as well as gasification of the pure substances were considered.

**EXPERIMENTAL SECTION**

**Pilot Plant.** DFB gasification was developed at the Vienna University of Technology in the 1990s for the thermal conversion of biomass, with the aim of producing a high-quality product gas. Steam is therefore used as the gasification agent, yielding a product gas with a high hydrogen content (>40%), low nitrogen content (<2%), and a moderate LCV of 12–14 MJ Nm⁻³ when wood is gasified. The DFB process was demonstrated successfully in 2001 in Güssing (Austria), with the first industrial-sized gasification plant of its type (8 MW fuel input); further information can be found in refs 16 and 17. Several other gasifiers based on DFB technology followed by now. The product gas of the DFB gasifiers is typically used in gas engines for heat and power production. Because of steam gasification, the product gas is also well-suited for the synthesis of gaseous and liquid fuels, such as synthetic natural gas, hydrogen, Fischer–Tropsch diesel, and mixed alcohols, which is investigated using the product gas from DFB gasifiers. The first industrial DFB plant producing synthetic natural gas is realized in Gothenburg (Sweden) in the GobiGas project.

At the Vienna University of Technology, a 100 kW pilot plant is operated for scientific purposes. Similar in design to industrial DFB gasifiers, the pilot plant is an essential tool for the evaluation of new feedstock, catalytic bed materials, and further development of the DFB gasifier. A schematic illustration of the pilot plant is shown in Figure 1.

The DFB gasification system comprises two separate reactors that are thermally connected by circulating bed material. In the gasification reactor, biomass is gasified with steam. Some of the ungasified char remains and is transported to the combustion reactor with the circulating bed material. Char is then combusted with air in the combustion reactor. Here, the bed material is heated, separated from the flue gas, and returned to the gasification reactor to deliver the energy needed for gasification. The DFB system yields two different gas streams: product gas and conventional flue gas.

The pilot plant provides fuel flexibility because of the presence of several gastight hoppers and screw feeders of different sizes. It is thus possible to feed material with a broad range of particle sizes and energy densities, to blend different materials for co-gasification, and to use different feed points located along the height of the reactor. Usually, the material is inserted directly into the fluidized bed via the screw conveyor of hopper 1. On top of the gasification reactor, a hopper for plastics has been installed, hopper 4, where the material is thrown onto the surface of the fluidized bed.

Olivein is the preferred bed material used in the DFB gasifier because it exhibits moderate tar cracking activity and has good mechanical stability. The bed material is provided by an Austrian manufacturer, Magnolithe GmbH. As different fuels are investigated in the pilot plant, the bed material is disposed of after each test run and fresh material is used, thus ensuring that all feedstock is tested in the same environment.

In the gasification reactor, a bubbling fluidized bed is created by superheated steam. The characteristic temperature of gasification is measured at the height of the screw feeder of hoppers 1 and 2 and is typically around 850 °C. Loop seals are installed to connect the gasification and combustion reactors; these seals are fluidized with steam to efficiently prevent gas leakage between the two reactors and to promote the transport of solids. In the combustion reactor, air is injected at two heights: primary air at the bottom of the reactor, where a dense fluidized bed is formed, and secondary air at a higher level to transport particles to the top of the reactor. Heat is generated via combustion in the combustion reactor, which determines the temperature in the gasification reactor. In addition to ungasified char from the feedstock, some fuel for combustion is inserted for temperature control. If no such fuel is added, the gasification temperature is moderated according to the energy demand of the gasification reactions and the amount of ungasified char available for combustion. In industrial gasifiers, ungasified char is transported with the bed material as well as tars and char separated from the product gas are fed to the combustion reactor as fuel for combustion. As a matter of simplicity, light fuel oil is used in the pilot plant for this purpose. The temperature difference between the combustion and gasification reactors amounts to 40–80 °C on average. Bed material is precipitated from the flue gas stream of the combustion reactor and returned to the gasification reactor, where it supplies heat for the endothermic gasification reactions. Downstream equipment is not included in Figure 1. A heat exchanger is arranged after the gasification reactor, where the product gas is cooled to about 250 °C before being sampled for analysis. Both product gas and flue gas are then mixed and combusted in a post-combustion chamber with air. A cyclone removes particles before the gas reaches the stack.

**Measuring Equipment.** A wide array of measuring equipment and automatic data recording were used at the pilot plant for data acquisition and process control. Temperatures of up to 1000 °C were measured with high-temperature thermocouples, while high-quality flow meters (Krohne) were employed for the adjustment of process media inputs, such as the fluidization agents, steam and air. Pressures were measured along the height of the reactors using pressure sensors relative to the atmosphere. The main product gas components, H₂, CO, CO₂, CH₄, and O₂, were analyzed online with a Rosemount NGA2000 device. C₅H₁₀, C₅H₁₂, C₆H₁₀, and the sum of gaseous C₂ and C₃ hydrocarbons were measured with a Syntech Spectras GC 955 gas chromatograph, with a sample taken every 20 min. In the course of this study, the gas chromatograph was replaced by a Perkin Elmar Arnel RGA101S, sampling every 15 min and analyzing N₂, C₂H₆, C₃H₈, and C₆H₁₀. In the flue gas from the combustion reactor, CO, CO₂, and O₂ were measured continuously via another Rosemount NGA2000 device.

Tar measurement was based on an impinger bottle method developed at the Vienna University of Technology. Similar to the conventional tar protocol, this method has been adapted for the analysis of the product gas from steam gasification. The tar absorbent was toluene. Because it is not miscible with water, dust, entrained char,

![Figure 1. Schematic illustration of the 100 kW gasification pilot plant.](Image)
water, and tar contents can be analyzed using the same sample. Two different methods of tar analysis were employed: gravimetric tar and GC−MS tar measurement. Gravimetric tars were weighed after vacuum evaporation of the solvent; these comprise mostly tars with a high molecular weight. A GC−MS device (gas chromatograph with a mass spectrometer) was used to measure the content of 50 different tar species of medium molecular weight in the product gas. The measurement ranges of the two techniques overlap, and therefore, both values are provided. A more detailed description of the tar measurement methodology is available in ref 31.

**Mass and Energy Balances with IPSEpro.** The process simulation tool IPSEpro was used for the evaluation and validation of the process data obtained during the experiments. IPSEpro offers stationary process simulation based on flow sheets and contains a comprehensive model library for gasification plants, which was developed at the Vienna University of Technology. The program is described in detail in ref 32. The mass and energy balances of the experimental runs were computed in IPSEpro. For this purpose, measured data from stationary operation of the pilot plant were used. An overdetermined equation system was formed that was solved by the method of least-squares; further information regarding this procedure can be found in ref 33. The reconciled solution best describes the actual operation of the pilot plant within the limits of the model.

**Feedstock.** In the present study, four different types of plastic material, including plastic residues as well as virgin polymers, were co-gasified with soft wood pellets: plastics from shredder light fraction (SLF), pellets made of selected plastic waste (MSW), PE regrind, and virgin PE. SLF material originates when end-of-life vehicles are derived from packaging waste, trashed plastic bags and waste bottles, which is a recycled product. PE regrind is usually made of used foils found in the selected plastics. PE consists only of volatiles, indicating that the constitutional hydrogen. In contrast, PE is comprised only of carbon and hydrogen hemicellulose that are mainly composed of carbon, oxygen, and hydrogen. In contrast, PE is comprised only of carbon and hydrogen arranged in long alkane molecules. Given that the constitutional repeating unit of PE is \( -(CH_2)_n \), its elemental composition can be estimated at around 85.6% carbon and 14.4% hydrogen, which is in good agreement with the elemental analysis obtained from the actual polymer used. PE regrind has a slightly higher carbon content than pure PE, which might stem from the regrinding process. The carbon content of SLF-plastics and MSW-plastics is also higher than that of wood, as polymers are generally rich in carbon because of their molecular structure. The LCV is strongly influenced by the water and oxygen contents, with low concentrations resulting in higher LCV, as is the case for plastic materials. However, the water content is only of minor importance for plastics because they do not absorb water to such a high extent as does biomass. The selected SLF-plastics and MSW-plastics contain about 10−12% ash, which is typical for waste materials, whereas the ash content of both the wood pellets and PE regrind is roughly 0.3%. Virgin PE contains virtually no ash. Inorganic compounds, such as nitrogen, sulfur, and chlorine, are only found in trace amounts in wood, PE, and PE regrind but amount to approximately 1% in SLF-plastics and MSW-plastics. Nitrogen, sulfur, and chlorine can be present as heteroatoms in polymers, such as nitrogen in polyamides, sulfur in vulcanized rubber, or chlorine in polyvinyl chloride (PVC). Determination of volatile content enables a first assessment of fuel devolatilization and gasification behavior. To eliminate the influence of the ash content, these volatile data are given on a dry and ash-free basis (daf). More volatile matter and less char are found in the selected plastics. PE consists only of volatiles, indicating that less char from these plastics is available for the combustion reactor.

<table>
<thead>
<tr>
<th> </th>
<th>wood pellets</th>
<th>SLF-plastics</th>
<th>MSW-plastics</th>
<th>PE</th>
<th>PE regrind</th>
</tr>
</thead>
<tbody>
<tr>
<td>LCV ( \text{kJ/kg} )</td>
<td>17458</td>
<td>31946</td>
<td>24092</td>
<td>43379</td>
<td>43270</td>
</tr>
<tr>
<td>water wt %</td>
<td>6.11</td>
<td>0.87</td>
<td>2.81 &lt;0.10 &lt;0.10</td>
<td></td>
<td></td>
</tr>
<tr>
<td>ash wt %, dry</td>
<td>0.29</td>
<td>10.67</td>
<td>12.47 &lt;0.10 0.36</td>
<td></td>
<td></td>
</tr>
<tr>
<td>C wt %, dry</td>
<td>50.23</td>
<td>65.00</td>
<td>54.16 85.84</td>
<td>87.09</td>
<td></td>
</tr>
<tr>
<td>H wt %, dry</td>
<td>6.04</td>
<td>7.95</td>
<td>7.32</td>
<td>14.07</td>
<td>12.42</td>
</tr>
<tr>
<td>( O^\circ ) wt %, dry</td>
<td>43.38</td>
<td>13.47</td>
<td>24.08 &lt;0.01 &lt;0.01</td>
<td></td>
<td></td>
</tr>
<tr>
<td>N wt %, dry</td>
<td>0.05</td>
<td>0.93</td>
<td>0.94</td>
<td>0.09 &lt;0.05</td>
<td></td>
</tr>
<tr>
<td>S wt %, dry</td>
<td>0.005</td>
<td>0.31</td>
<td>0.21 &lt;0.005 0.01</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cl wt %, dry</td>
<td>0.003</td>
<td>1.67</td>
<td>0.82 &lt;0.005 0.07</td>
<td></td>
<td></td>
</tr>
<tr>
<td>volatiles wt %, daf (^a)</td>
<td>86.7</td>
<td>89.2</td>
<td>90.2 &gt;99</td>
<td>99.5</td>
<td></td>
</tr>
</tbody>
</table>

\(^a\)Calculated by difference to 100%. \(^b\)daf = dry and ash-free basis.
The main focus of the investigation was the change in gasification behavior resulting from the different mixing ratios. As a consequence, the main parameters of the DFB pilot plant were kept constant while using one type of plastic material. Important data from stationary operation of the pilot plant for all test runs were averaged and presented in Table 3 (wood, SLF-plastics, and MSW-plastics) and in Table 4 (PE regrind and PE). The nominal fuel power of the DFB pilot plant amounts to 100 kW, which was applied for the gasification of wood, SLF-plastics, and MSW-plastics. When 100% SLF-plastics was gasified, the total fuel input was 89 kW because of the limitations of the feeding system, it was not possible to further increase the mass flow. Fuel power was also reduced for PE and PE regrind, because these polymers are likely to generate product gas with a high calorific value. In the pilot plant, product gas is combusted in the post-combustion chamber of the pilot plant together with the flue gas after analysis. Because the fuel power of the post-combustion chamber is limited, the fuel power of the gasification reactor was reduced to 90 kW in this case. The characteristic temperature in the gasification reactor was kept constant at 850 °C (±3 °C) in all experiments. The plastic materials were generally fed from hopper 4 (Figure 1). For the gasification of the mixtures containing 25 and 100% SLF-plastics, SLF-plastics were fed directly into the fluidized bed from hopper 2. Wood pellets were always discharged from hopper 1 and fed into the fluidized bed. In a recently published study at the DFB pilot plant, the effects of in- and on-bed feeding were investigated using, among other materials, a mixture of 50% wood pellets and 50% SLF-plastics (on the basis of energy). Wood pellets are fed directly into the fluidized bed during in- and on-bed feeding of SLF-plastics. Almost no differences are found in product gas composition when the feeding position is changed. The GC–MS and gravimetric tars also remain constant. Because SLF-plastics devolatilize rapidly, fewer fuel particles remain in the bed to be gasified there. The gas-phase reactions mostly take place in the freeboard section with limited contact to bed material. The constant steam of wood pellets into the bed increases the turbulence in the bed and reduces the impact of the different feed points for SLF-plastics. It is found that changes in the feed point of SLF-plastics do not influence the gasification process significantly.

A total of 100 kg of olivine was used as bed material in the DFB reactor, with a particle size distribution ranging from 0.4 to 0.6 mm. The steam/carbon ratio describes the relationship between the mass flows of the fluidization steam and the fuel water and the mass flow of fuel carbon according to eq 1. It was in the range of 1.8 (kg/kg) for SLF-plastics and MSW-plastics and was raised to 2.0 (kg/kg) for 100% SLF-plastics and 2.3 (kg/kg) for PE regrind and PE. Although the amount of fluidization steam for mono-gasification of plastic materials should be lowered because of the high carbon content of the polymers, a minimum amount is required to maintain circulation and sufficient fluidization of the fluidized bed. For the gasification of wood, a S/C ratio of 2.1 (kg/kg) was chosen to fit with all other gasification tests. The combustion reactor was typically fluidized with primary and secondary air in the range of 54 N m³/h, with primary air amounting to about 9% of secondary air. The combustion temperature exceeded the gasification temperature by a maximum of about 80 °C.

Product Gas. Different mixing ratios of biomass and plastics influence the product gas composition, which is shown in Figures 2 and 3. For H₂ and CO production, these two figures display not only measured values but also a linear interpolation based on experimental data for pure materials. The lines do not appear as linear relationships here, because the energy-based mixing ratios and not the mass-based ratios are used on the x axis in Figure 2. The dashed lines present the values of the linear interpolation, where the H₂ or CO production from the mixture is calculated from the results of gasification of the pure feedstock according to their mass fraction. More H₂ was generated from mono-gasification of PE regrind and SLF-plastics than from wood pellets alone. The H₂ concentration remains almost constant, when virgin PE was used. When mixtures were gasified, nonlinear effects occurred but no general trend was observed in terms of H₂ formation for the different materials. While linear interpolation overestimated the H₂ yield from SLF-plastic and MSW-plastic mixtures, more H₂ was produced from PE and PE regrind than calculated. Interestingly, PE and PE regrind behaved differently, despite varying the least in elemental composition. This discrepancy was eventually traced to the presence of impurities in the employed PE regrind material.

The concentration of CO decreased strongly when only plastics or any mixture of plastics and wood were gasified. Nonlinear effects are also visible but not as much as for H₂.

Table 2. Overview of the Co-gasification Experiments

<table>
<thead>
<tr>
<th>share of plastics (on the basis of energy)</th>
<th>25%</th>
<th>50%</th>
<th>75%</th>
<th>100%</th>
</tr>
</thead>
<tbody>
<tr>
<td>SLF-plastics</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>PE regrind</td>
<td>x</td>
<td>x</td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td>PE</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>MSW-plastics</td>
<td>x</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 3. Operational Data (Part 1)

<table>
<thead>
<tr>
<th>share of plastics (on the basis of energy)</th>
<th>wood pellets</th>
<th>SLF-plastics</th>
<th>SLF-plastics</th>
<th>SLF-plastics</th>
<th>SLF-plastics</th>
<th>MSW-plastics</th>
</tr>
</thead>
<tbody>
<tr>
<td>fuel input</td>
<td>%</td>
<td>0</td>
<td>25</td>
<td>50</td>
<td>75</td>
<td>100</td>
</tr>
<tr>
<td>steam/carbon ratio</td>
<td>kg</td>
<td>97</td>
<td>98</td>
<td>98</td>
<td>99</td>
<td>89</td>
</tr>
<tr>
<td>feeding position*</td>
<td>bed</td>
<td>bed</td>
<td>top</td>
<td>top</td>
<td>bed</td>
<td>top</td>
</tr>
<tr>
<td>gasification temperature</td>
<td>°C</td>
<td>856</td>
<td>855</td>
<td>852</td>
<td>850</td>
<td>851</td>
</tr>
<tr>
<td>combustion temperature</td>
<td>°C</td>
<td>883</td>
<td>897</td>
<td>895</td>
<td>904</td>
<td>921</td>
</tr>
<tr>
<td>total steam fluidization (gasification reactor)</td>
<td>kg/h</td>
<td>18.9</td>
<td>15.0</td>
<td>14.3</td>
<td>13.7</td>
<td>13.0</td>
</tr>
<tr>
<td>total air fluidization (combustion reactor)</td>
<td>N m³/h</td>
<td>55.3</td>
<td>55.1</td>
<td>52.7</td>
<td>54.6</td>
<td>53.7</td>
</tr>
</tbody>
</table>

*Top, hopper 4; bed, hopper 2 (in Figure 1).
Similar to CO, the concentration of CO$_2$ also decreased. A slight decline was apparent during co-gasiﬁcation of SLF-plastics and MSW-plastics, with a steeper decline for PE and PE regrind. These decreases in CO and CO$_2$ are mainly related to the reduced amount of oxygen in the feedstock. None at all was present when 100% PE and PE regrind were gasiﬁed. In this case, CO and CO$_2$ were formed only as a result of reactions with steam. The results presented here are in good agreement with a similar gasiﬁcation study using a steam-blown bubbling bed gasiﬁer. Pinto et al.$^{10}$ also observed increased production of H$_2$ and a decline in CO and CO$_2$ with increasing proportions of PE in mixtures with pine.

CH$_4$ and C$_2$H$_4$ are typical decomposition products of polymers produced in DFB gasiﬁers.$^{37}$ During mono-gasiﬁcation of the plastic materials in the present study, considerably high concentrations of CH$_4$ and C$_2$H$_4$ were measured. Whereas the increase in CH$_4$ was almost linear for SLF-plastics, for PE and PE regrind this increase was nonlinear. Gasiﬁcation of the 75% PE regrind mixture produced signiﬁcantly lower CH$_4$ and C$_2$H$_4$ concentrations than expected. In the product gas, also trace amounts of C$_3$H$_6$ and other gaseous C$_3$ and C$_4$ hydrocarbons were detected. The C$_3$H$_6$ content ranged from 0.1 to 0.7 vol % and behaved similarly to C$_2$H$_4$ with increasing proportion of plastics. The concentrations of C$_3$H$_8$ and other gaseous C$_3$ and C$_4$ hydrocarbons were even smaller.

The LCV of the product gas was calculated on the basis of product gas composition; the tar and char contents of the product gas were not considered in this calculation. Because CH$_4$ and C$_2$H$_4$ have high LCV, any change in the latter with the proportions of the various plastics will behave similar to the variation in CH$_4$ and C$_2$H$_4$ production. Accordingly, the LCV increased when more plastic materials were used in the feedstock mixtures.

Biomass and polymers interact during steam gasiﬁcation, resulting in nonlinear changes to product gas composition. According to the pyrolysis studies carried out by Jakab and co-workers,$^{3,4}$ the presence of biomass char has a signiﬁcant eﬀect on product distribution. It inﬂuences both inter- and intramolecular H transfer during the radical decomposition mechanism of polymers. The eﬀect of char on polymer decomposition was also found to depend upon the type of

### Table 4. Operational Data (Part 2)

<table>
<thead>
<tr>
<th>Share of plastics (on the basis of energy) %</th>
<th>PE</th>
<th>PE</th>
<th>PE regrind</th>
<th>PE regrind</th>
<th>PE regrind</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel input kW</td>
<td>90</td>
<td>100</td>
<td>25</td>
<td>50</td>
<td>75</td>
</tr>
<tr>
<td>Steam/carbon ratio kg/kg</td>
<td>2.3</td>
<td>2.3</td>
<td>2.3</td>
<td>2.3</td>
<td>2.3</td>
</tr>
<tr>
<td>Gasiﬁcation temperature °C</td>
<td>854</td>
<td>852</td>
<td>850</td>
<td>850</td>
<td>850</td>
</tr>
<tr>
<td>Combustion temperature °C</td>
<td>893</td>
<td>895</td>
<td>918</td>
<td>915</td>
<td>915</td>
</tr>
<tr>
<td>Total steam ﬂuidization (gasiﬁcation reactor) kg/h</td>
<td>17.0</td>
<td>15.0</td>
<td>18.1</td>
<td>16.7</td>
<td>16.1</td>
</tr>
<tr>
<td>Total air ﬂuidization (combustion reactor) N m$^3$/h</td>
<td>56.3</td>
<td>54.3</td>
<td>53.4</td>
<td>52.9</td>
<td>53.1</td>
</tr>
</tbody>
</table>

“Top = hopper 4; bed = hopper 2 (in Figure 1).”

Figure 2. H$_2$ and CO in dry product gas (measured and calculated).
polymer. Ahmed et al.\textsuperscript{9} determined an alternate influence of wood and plastics and postulated that wood chips absorb PE volatiles and hence promote steam reforming of the volatiles. PE radicals might act as H donors that stabilize the radicals formed by wood chips. Co-gasi
\textsuperscript{f}cation yielded lower CH\textsubscript{4} and C\textsubscript{2}H\textsubscript{4} contents than expected but higher concentrations of CO and H\textsubscript{2}. The change in gas composition is indicative of enhanced steam reforming. These results could possibly be related to the presence of char from biomass.

Mass and energy balances of the gasification test were calculated on the basis of measured values using IPSEpro. The accuracy of these calculations depends upon that of the measured data. In the case of plastic materials, the tar content has a significant influence. As a result of missing or failed tar measurements, no reliable mass and energy balance values were available for mixtures containing 75\% SLF-plastics and 100\% PE regrind. Further information can be found in the following section (Tar Formation).

The generation of product gas (dry) and water conversion for each of the mixtures is illustrated in Figure 4. For sound comparison, volume flows were converted into those for a fuel input of 100 kW. The product gas volume flow decreased when mixtures of wood pellets and plastics and plastics only were gasified, although this decline was less strong than expected from mono-gasi
\textsuperscript{f}cation. For example, the 50\% PE mixture produced considerably more gas than gasi
\textsuperscript{f}cation of 100\% PE. However, when mixtures containing PE regrind were gasified, a larger volume of dry product gas was generated than that observed during wood gasi
\textsuperscript{f}cation. Mastellone et al. investigated co-gasi
\textsuperscript{f}cation of coal, plastic waste, and biomass in a bubbling fluidized-bed gasifier and compared the relative influence of the different fuels. Plastics are found to increase the specific gas production.\textsuperscript{12} If the values in Figure 4 were expressed in terms of the mass of fuel input, a similar pattern would be observed. Mixtures containing plastics yielded more product gas compared to wood.

Figure 3. CO\textsubscript{2}, CH\textsubscript{4}, C\textsubscript{2}H\textsubscript{4}, and LCV of dry product gas.

Figure 4. Product gas volume flow (dry) and water conversion.
Water conversion, $X_{\text{H}_2\text{O}}$, describes how much water is consumed by the gasification reactions and is defined as the ratio of water consumption to water supply (fluidization steam and fuel bound water), as in eq 2. The steam mass flow and the amount of fuel water were measured values. The amount of water consumed by the gasification reactions resulted from the mass and energy balance calculated with IPSEpro based on measured data from the experiment. Water conversion summarizes the effect of several gasification reactions, including the steam gasification of solid carbon ($\text{C} + \text{H}_2\text{O} \rightarrow \text{CO} + \text{H}_2$), the water–gas shift reaction ($\text{CO} + \text{H}_2\text{O} \leftrightarrow \text{CO}_2 + \text{H}_2$), and steam-reforming reactions of hydrocarbons [$\text{C}_m\text{H}_n + m\text{H}_2\text{O} \rightarrow m\text{CO} + (m + 0.5n)\text{H}_2$]. In general, the water conversion correlates with the dry product gas flow, because at high levels of water conversion, more water is converted into combustible product gas compounds. The lowest water conversion values were determined for 100% wood and 100% PE. Co-gasification of plastics and wood resulted in an increase in water conversion, a pattern that also correlates with the deviation from the interpolation based on pure substances. As is apparent in Figure 2, larger volumes of H2 and CO than expected were measured when PE or PE regrind were part of the mixtures. The water conversion of the mixtures was also higher. Apparently, the steam reforming reactions were enhanced. In contrast, mixtures containing SLF-plastics and MSW-plastics behaved in a different manner. The water conversion increases continuously with an increasing share of SLF-plastics; the maximum is achieved at 100% SLF-plastics. Furthermore, CH4 increased almost linearly and less steeply compared to the other polymers. A recent study conducted at the 100 kW pilot plant investigated mono-gasification of polymers and polymer mixtures. This study found the water conversion to be greater when mixtures of different polymers were gasified. Most likely, more radicals of different types are available to interact with steam.37 SLF-plastics and MSW-plastics are comprised of a broad range of polymers. The water conversion in the present study was greater during mono-gasification of SLF-plastics compared to that of the mixtures. In contrast, the water conversion of 100% PE, which consists of only one type of polymer, was lower compared to that observed during co-gasification.

**Tar Formation.** Several co-gasification studies have shown that the presence of plastics in the fuel mixture increases the tar concentration in the product gas. This increase in the tar concentration is also accompanied by rising yields of CH4 and light hydrocarbons.11−13 Figure 5 summarizes the tar, dust, and char concentrations measured in the product gas during the experiments. The values are referred to the fuel power of the gasification reactor to remove the influence of different gas production. As mentioned previously, no tar measurement was available for the 75% SLF mixture, and the measurement failed for 100% PE regrind. Polymerization occurred in the tar sampling equipment, and thus, the particle separators and piping were rapidly clogged. Because it was not possible to dissolve all of the tar, which was stuck in the equipment, part of the sample had to be removed via combustion and, thus, could not be considered in the analysis. Because of this fact, the tar sample of 100% PE regrind was then omitted from further study.

Dust is comprised of inorganic solids, such as ash or entrained olivine particles. When SLF-plastics and MSW-plastics were gasified, the dust content increased, which is typical for waste fuels because they contain more ash.
Interestingly, the dust content decreased during gasification of 100% SLF, potentially indicating that the ash remained in the bed instead of being carried out. A possible explanation therefore is that 100% SLF-plastics was fed directly into the fluidized bed. In combination with lower gas production compared to other SLF-mixtures, the ash is more likely to remain in the bed. According to the fuel analysis in Table 1, PE regrind contained only trace amounts of ash; dust from PE regrind was therefore more likely to contain olivine particles. Combustible solids contained in the product gas are summarized as char. In general, less char is available from highly volatile fuels. Here, the char content decreased with an increasing share of PE. More char was entrained when mixtures containing SLF-plastics were gasified, which might be related to the increased gas formation for the mixtures but also to charring materials present in SLF-plastics.

Concentrations of gravimetric and GC−MS tars rose nonlinearly with an increasing proportion of plastics in the fuel mixture, similar to the change in the CH₄ content. The concentrations of gravimetric and GC−MS tars changed in a comparable way. Gravimetric tars comprise mostly tars with a high molecular weight, whereas GC−MS tars have medium molecular weight. The measurement ranges of the two techniques overlap, and therefore, both values are provided. Typically, the concentration of GC−MS tars is higher than the concentration of gravimetric tars. The difference in the tar concentration between the 50 and 100% PE mixtures was large. The tar content yielded from 75% PE regrind was also comparably lower than tar from 100% PE and 100% SLF. The latter results could provide an estimate for tar formation from the gasification of 100% PE regrind. When the mixture containing 25% SLF-plastics was gasified, the material was fed directly into the fluidized bed together with the wood pellets. During gasification of 100% SLF-plastics, the material was also fed directly into the bed. In this way, the volatiles were able to flow through the fluidized bed and can have longer contact with the catalytic bed material than during on-bed feeding. The tar content could be lower during in-bed feeding. However, a study on in-bed and on-bed feeding at the DFB pilot plant showed that changes in the feeding position do not influence the product gas composition and tar formation significantly, when a mixture of 50% wood pellets and 50% SLF-plastics (on the basis of energy) was used. Because devolatilization of plastics occurs rapidly, the gas-phase reactions take place mainly in the freeboard region with limited contact with the bed material. The constant stream of wood pellets into the bed increases the

Table 5. Substance Groups for GC−MS Tar Classification

<table>
<thead>
<tr>
<th>group</th>
<th>substance</th>
</tr>
</thead>
<tbody>
<tr>
<td>phenols</td>
<td>phenol, 2-methylphenol, 4-methylphenol, 2,6-dimethylphenol, 2,4-dimethylphenol, 2,5-dimethylphenol, 3,5-dimethylphenol, 2,3-dimethylphenol, 3,4-dimethylphenol, 2-methoxy-4-methylphenol, and catechol</td>
</tr>
<tr>
<td>furans</td>
<td>benzo[b]furan, 2-methylbenzofuran, and dibenzofuran</td>
</tr>
<tr>
<td>aromatics</td>
<td>phenylacetylene, styrene, mesitylene, 1H-indene, and 1-indanone</td>
</tr>
<tr>
<td>naphthalenes</td>
<td>naphthalene, 2-methylanthracene, and 1-methylnaphthalene</td>
</tr>
<tr>
<td>PAHs</td>
<td>biphenyl, acenaphthylene, acenaphthene, fluorene, anthracene, phenanthrene, 4,5-methylphenanthrene, 9-methylanthracene, fluoranthenes, pyrene, benz[a]anthracene, chrysene, benz[k]fluoranthene, benz[k]fluoranthene, benzo[a]pyrene, benzo[g,h,i]perylene, indeno[1,2,3-cd]pyrene, and dibenz[a,k]anthracene</td>
</tr>
</tbody>
</table>
turbulence in the bed and reduces the impact of the different feed points for SLF-plastics.36 When the results of 25% SLF-plastics are compared to 50% SLF-plastics in this work, no clear influence of the feeding position on the tar content is apparent either. The presence of wood char seems to have more impact.

The lower tar formation from 100% SLF-plastics compared to PE could be attributed to in-bed feeding but also to different types of radicals available from the polymer mixture in SLF-plastics. These radicals interact with steam37 and enhance steam reforming of hydrocarbons.

Co-gasification of biomass and plastics resulted in a nonlinear increase in tar formation in the DFB gasifier. This is also reported by Berrueco et al.8 and Mastellone et al.12 These authors explained their results by the presence of wood char, as also demonstrated in the experiments by Boroson et al.38 A total of 50 substances in tar were identified by GC−MS analysis, which provides deeper insight in the nature of tars. For reasons of clarity and comprehensibility, these substances are grouped according to their chemical functionality based on ref 31 in Figure 6. The constituents of the substance groups are listed in Table 5. Phenols and furans contain oxygen and are typically associated with tars from wood gasification. These two groups are shown together in Figure 6. Tars from co-gasification of wood and plastics contained less phenols and furans. This decrease was once again nonlinear and steeper than expected. Tars from plastic materials did not contain any oxygenated compounds. The share of aromatic compounds also decreased, and that of polycyclic aromatic hydrocarbons (PAHs) remained more or less constant; however, there was no clear trend of how the concentrations of aromatic compounds and PAHs change with an increasing share of plastics in the mixtures. Naphthalene is the most important tar compound generated in the DFB gasification system. Typically, about 25% of the tar from wood consists of naphthalene. The substance groups of naphthalenes also increased with an increasing proportion of plastics. In conclusion, the composition of GC−MS tars is influenced by the fuels used in the gasification process. This is in agreement with the findings by Ruoppolo et al.,11 who compared the co-gasification of 20 wt % PE and pine with pure pine gasification. The authors used an air- and steam-blown bubbling bed gasifier at 780 °C with a Ni-based catalyst and also provided some information as to the composition of the produced tars. When 20 wt % PE is added to pine, tar composition changes markedly. The share of phenols, aromatic compounds, and naphthalenes decrease, whereas the proportion of PAHs increases to more than two-thirds of the total tar content. When wood pellets are gasified, the most important single substance is naphthalene. In the mixture containing PE, phenanthrene predominated.

**Energy Distribution in the DFB Gasification System.**

The studied DFB gasification system consists of an allothermal gasification reactor and a combustion reactor, which provides the energy for gasification. The circulating bed material serves as an energy carrier. In the gasification reactor, residual char from gasification is combusted. To control the temperature in the gasification reactor, some other fuel is fed to the combustion reactor. In industrial-scale DFB gasifiers, tars and char contained in the product gas and other combustible streams are recycled to the combustion reactor. In the 100 kW pilot plant, these streams are replaced by light fuel oil. Thermal losses to the environment are much higher in the pilot plant than in any industrial plant. In large-scale DFB gasifiers, heat losses are more or less negligible because of the quality of the insulation and the more advantageous surface/volume ratio, and hence, less fuel for combustion is required. To be able to obtain useful data from the pilot plant for industrial-sized plants with respect to the fuel demand of the combustion reactor, the...
measured values were corrected for heat losses of the pilot plant, as in eq 3.

specific power of combustion

\[
SPC = \frac{P_{\text{fuel,combustion}} - P_{\text{losses}}}{P_{\text{feedstock}}} \tag{3}
\]

The amount of residual char, the specific power of combustion, and the specific power of gasification were selected to describe the energy balance of the DFB gasification system. Residual char from gasification is transported to the combustion reactor together with the circulating bed material. For calculation of the mass and energy balances, the composition of the char is required. Because the composition of the residual char could not be measured at the pilot plant, the following values determined in earlier work were used for wood char: 3.8% H, 14% O, and 82.2% C. A previous study investigating mono-
determined in earlier work were used for wood char: 3.8% H, 14% O, and 82.2% C. A previous study investigating mono-
gasification of polymers suggested that char is transported to the combustion reactor, despite the highly volatile nature of polymers. Most likely, carbonaceous deposits are formed on olivine particles, which are assumed to be similar to polyaromatic hydrocarbons with 8% H and 92% C. For co-
gasification, char composition was calculated on the basis of the mass ratio of wood to plastics. It was also assumed that no char was transported back to the gasification reactor. In Figure 7, the flow of the residual char is referred to the fuel input of the gasification reactor.

The circulating bed material delivers heat to the gasification reactor. This heat transfer can be described in terms of the temperature difference between the incoming and outgoing bed material. Part of the heat is used to cover the thermal losses of the gasification reactor, \( P_{\text{losses, gasification}} \), as well as to heat the fluidization steam from approximately 250 °C to the gasification temperature of 850 °C, \( P_{\text{heat,steam}} \). The remaining power is consumed by the gasification reactions for the formation of the hot product gas, as expressed in eq 4.

specific power of gasification

\[
SPG = \frac{P_{\text{bed, in}} - P_{\text{bed, out}} - P_{\text{losses, gasification}} - P_{\text{heat, steam}}}{P_{\text{feedstock}}} \tag{4}
\]

The amount of residual char decreased when the proportion of plastics increased in the mixtures. The specific power of combustion rose markedly and nonlinearly with an increasing share of plastics. The interpolation based on pure substances underestimated the required specific power of combustion. The maximum specific power of combustion was necessary for gasification of the mixture containing 75% PE regrind, during which the highest water conversion rate occurred. The specific power of gasification decreases with an increasing share of plastics, which is most likely a property of the material. The waste materials, SLF-plastics and MSW-plastics, required less power of combustion than PE regrind and PE. An important difference between these waste materials and PE polymers is the oxygen content. Oxygen bonds are very reactive, which is also a possible explanation for the lower power of gasification for the waste materials.

■ FLUID DYNAMICS IN THE GASIFICATION REACTOR

Product gas yield depends upon the composition of the fuel mixture and the operating conditions, as illustrated in Figure 4. Figure 8 shows the fluidization number, \( U/U_{\text{mf}} \) for each of the studied sample mixtures. During the gasification tests, product gas velocity ranged from 0.38 to 0.66 m/s in the gasification reactor, while the averages of the gas residence time varied between 2.5 and 4 s. The minimum fluidization velocity \( U_{\text{mf}} \) was calculated for olivine particles and amounted to 0.16 m/s. The fluidization number declined with an increasing proportion of plastics in the fuel mixtures. Because actual operation values were used in this calculation, the results also reflect differences in load and S/C ratio. For instance, the lowest values were observed for SLF-plastics with a S/C ratio of 1.8 (kg/kg). When 100% plastic materials were gasified, low fluidization numbers were achieved, corresponding to the onset of the bubbling bed regime. This shows that the bubbling bed was less active during the gasification of these samples.

At the DFB pilot plant, feedstock can be inserted into the gasification reactor at different heights. During the gasification tests, polymers were mostly fed from the top of the gasifier, while wood pellets were fed directly into the fluidized bed. The feeding positions used in all of the test runs are given in Tables 3 and 4. Plastic particles are discharged from the screw conveyor at the top of the gasification reactor. During their fall, the plastic particles heat, melt, and devolatilize. Terminal velocity was calculated for the smallest and lightest PE particles to check whether the particles really fell in the countercurrent product gas flow. According to these calculations, the terminal velocity of PE particles amounted to 14 m/s; it took the particles 0.1 s to fall through the freeboard and land in the bubbling bed. Wood pellets were fed directly into the fluidized bed, which is the part of the reactor characterized by the highest heat-transfer rates. Here, the pellets heat, dry, and devolatilize. The emerging volatiles form bubbles that lift the pellets up in the bubbling bed. Because of devolatilization, the splash zone is more active when wood is fed into the bed. The bed material that returns from the combustion reactor is then thrown onto the surface of the fluidized bed, which enhances intermixing of fuel particles, gases, and bed material.

Plastic materials have a high content of volatile matter, especially PE and PE regrind. Therefore, their polymers are likely to react mainly in the freeboard and splash zone. When 100% polymer samples were gasified, the splash zone was less active compared to during gasification of mixtures, because the gas bubbles of the volatiles from wood were absent and, thus, less gas was produced. However, the polymers still entered the bed because they contributed to char combustion in the combustion reactor.
The temperature profile of the gasification reactor appears in Figure 9. A total of five temperature measurement points were positioned along the height of the reactor and show hot and cold spots. The characteristic gasification temperature (shown in Tables 3 and 4) was measured at a height of 0.8 m, where the wood pellets were inserted into the bed. During gasification of wood, the temperatures deep in the bubbling bed were lower. This indicates that heat was consumed by the gasification reactions taking place there. The bed temperature increased with an increasing share of PE in the fuel mixture; therefore, the gasification reactions moved upward to higher levels of the bubbling bed. The highest temperature was achieved in the lower part of the freeboard, where the hot bed material returned from the combustion reactor. In the upper part of the freeboard, the temperature decreased with an increasing height, mainly because of thermal losses from the pilot plant. Figure 9 shows the beneficial effects of co-gasification of biomass and plastics. Because of the fuel mixture, gasification took place deep within the bed and inside the splash zone. The splash zone was also more active because of the bubbles generated by the volatiles from wood. Contact between gas and bed material was therefore enhanced, which is decisive in catalytic tar reduction. This is confirmed by the lower tar concentrations observed during co-gasification. Co-gasification also resulted in enhanced steam reforming and, therefore, higher water conversion rates.

Figure 9. Temperature profile along the height of the gasification reactor during gasification of PE mixtures.

**CONCLUSION**

Co-gasification of biomass and plastics was investigated in the 100 kW DFB pilot plant at the Vienna University of Technology. A total of four different plastic materials, including both waste materials and virgin polymers, were blended with soft wood pellets. The proportion of plastics was varied in a broad range; each material was mixed with wood in a 50:50 ratio (on the basis of energy), and two materials were also tested in several mixing ratios ranging from 0 to 100% plastics.

Product gas composition was considerably influenced by co-gasification. Significantly, these changes were nonlinear, and therefore, gas composition could not be accurately predicted on the basis of mono-gasification of the materials only. More CO and CO₂ were measured in the product gas from co-gasification than would have been expected from linear interpolation of the pure substances, whereas H₂ production was either underestimated or overestimated depending upon the plastic material. Smaller amounts of CH₄ and C₂H₆ were formed than expected, which was also the reason for the lower LCV of the product gas. The tar content in the product gas was also lower than presumed. With an increasing share of plastics in the fuel mixtures, the composition of the GC–MS tars changed: less phenols and furans and more naphthalenes were formed. Co-gasification of biomass and plastics yielded more product gas than expected, largely because of the enhanced reactions with steam and, therefore, increased water conversion. This was also the reason for the deviation of product gas composition from the results of linear interpolation. It also matches with the observation that apparently more endothermal reactions took place during co-gasification, because the steam reforming of hydrocarbons was enhanced. As a consequence, the fuel demand for combustion increased nonlinearly, while less char from the feedstock was available for combustion. When biomass was gasified in the DFB gasifier, gasification occurred throughout the height of the fluidized bed. Mono-gasification of plastics mainly occurred in the upper part of the bubbling fluidized bed and the freeboard. Because of devolatilization of wood pellets in the fluidized bed, more bubbles were formed in the bed. In comparison to mono-gasification of plastics, a more active splash zone was established during co-gasification. Because of the more active splash zone, the contact between gas and bed material was prolonged, which is crucial for catalytic tar removal. Co-gasification of wood and plastic materials also had other beneficial effects: more radicals of different types were available from fuel mixtures that interacted with each other and with the fluidization steam. In addition, the presence of wood char had a positive effect on polymer decomposition, steam reforming, and tar reduction. Co-gasification of plastics and biomass significantly reduced the tar formation compared to mono-gasification of plastics, where the high tar formation is problematic for industrial applications. Co-gasification with biomass is therefore a suitable way to use plastic materials in DFB gasifiers.

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**Notes**
The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

The authors thank the “Gasification and Gas Cleaning” working group and the “Test Laboratory for Combustion Systems” at the Institute of Chemical Engineering, Vienna University of Technology, for their support. Many thanks also go to Borealis Polyolefine GmbH, Austria, for providing the virgin polymers and AVE Österreich GmbH, Technische Behandlungssysteme (TBS)GmbH, and the University of Ljubljana for providing the polymer residues. Bioenergy2020+ is funded within the Austrian COMET program managed by the Austria Research Promotion Agency (FFG). The financial support given by the funding association FFG and the industrial partners Magna International AG, Repotec Umwelttechnik GmbH, and Biomasse-Kraftwerk-Güssing GmbH is gratefully acknowledged.
NOMENCLATURE

Abbreviations

- DFB = dual fluidized bed
- DTG = differential thermogravimetry
- GC–MS = gas chromatography–mass spectrometry
- LCV = lower calorific value
- MSW = municipal solid waste
- OECD = Organization for Economic Co-operation and Development
- PE = polyethylene
- PP = polypropylene
- PS = polystyrene
- S/C = steam/carbon ratio
- SLF = shredder light fraction
- SPC = specific power of combustion
- SPG = specific power of gasification
- TGA = thermogravimetric analysis
- TG–MS = thermogravimetry–mass spectrometry

Symbols

- $P$ = power (kW)
- $U$ = velocity (m/s)
- $X$ = conversion (%)

Subscripts

- $\text{bed, in}$ = bed material going into the gasification reactor
- $\text{bed, out}$ = bed material leaving the gasification reactor
- $\text{carbon, feedstock}$ = carbon in the feedstock
- $\text{feedstock}$ = feedstock fed into the gasification reactor
- $\text{fuel, combustion}$ = fuel fed into the combustion reactor
- $\text{heat, steam}$ = energy to superheat fluidization steam
- $\text{H}_2\text{O, consumed}$ = water consumed in gasification reactions
- $\text{H}_2\text{O, fluidization}$ = fluidization steam
- $\text{H}_2\text{O, feedstock}$ = fuel-bound water
- $\text{losses}$ = thermal losses
- $\text{losses, gasification}$ = thermal losses of the gasification reactor
- $\text{mf}$ = minimum fluidization

REFERENCES