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# Gasification of plastic residues in a dual fluidized bed gasifier – Characteristics and performance compared to biomass

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## Abstract

Plastic residues are gasified in a dual fluidized bed gasifier. Two different types of plastic residues are used: shredder light fraction from end-of-life vehicles and pellets made of selected plastics from municipal solid waste. Different mixtures of plastic residues and soft wood pellets ranging from 0-100%, as well as different steam-to-carbon ratios are tested. During the gasification experiments producer gas and flue gas composition are measured, as well as tars, entrained dust, and char. In order to evaluate the results, experimental data of the test runs with plastic residues is compared to gasification of wood pellets. It is found that increasing share of plastics in the fuel mixture leads to higher concentrations of CH<sub>4</sub>, C<sub>2</sub>H<sub>4</sub> and other light hydrocarbons in the producer gas. Producer gas with higher calorific value is yielded. The tar and dust content of the producer gas increases, whereas the amount of entrained char decreases. Mixtures of plastics and wood cause changes in the producer gas composition in a non-linear way, which indicates interaction of both feedstock. An increasing demand of auxiliary fuel is observed, which originates from the lower char content in plastic residues.

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## 1. Introduction

Biomass gasification has been studied in detail during the last decades and shows remarkable progress. The dual fluidized bed gasification system is now commercially available. At the end of this year four industrial plants with capacities ranging from 8.5 to 15 MW will be in operation in Europe. These gasifiers are designed for wood chips from forestry as feedstock. However, alternative feedstock for biomass gasifiers is in demand by operators and manufacturers. The use of e.g. plastic residues in biomass gasifiers can increase the feedstock flexibility of the gasifier and might also offer some economic advantages. Research is necessary to determine the suitability of new feedstock. It is essential to understand the influence of the feedstock on the gasification behavior, as well as the effects on the whole process chain.

Several studies on gasification of plastics are available in literature for gasifiers of different types and scale. Pohorely et

al. [1] gasified PET and coal in a bubbling fluidized bed gasifier using 10% oxygen in bulk nitrogen as gasification agent. It is found that PET reacts much faster than coal, but it also increases the tar content of the producer gas. It is assumed that tars are formed from volatile matter, which leave the gasifier unreacted. Mastellone et al. [2] investigated air gasification of recycled PE, coal and wood in a lab-scale bubbling fluidized bed gasifier. They reported synergy effects, when mixtures are gasified, whereas PE increases the syngas yield and the lower calorific value of the producer gas. Wood enforces dust and char entrainment and reduces the tar content. Pinto et al. [3] tested different mixtures of PE and pine wood in a steam blown bubbling fluidized bed gasifier at lab-scale. They observed that H<sub>2</sub> increases, whereas CO decreases with increasing share of PE in the feed mixture. Arena et al. [4] operated a bubbling fluidized bed gasifier at pilot scale with air fluidization. They investigated different waste derived

plastics and reported high  $H_2$  and CO concentrations during gasification of polyolefin wastes. Besides investigations in laboratory scale, there are also several industrial gasifiers (e.g. Lahti, Rüdersdorf, Grève in Chianti), where plastic residues have been used as feedstock. Mixtures of plastic residues and other fuels were gasified in air blown circulating fluidized beds, but no detailed studies on the gasification behavior of these plastic wastes are reported [5].

This paper presents the experimental results of gasification of selected plastic residues together with wood in a 100 kW gasification pilot plant. The aim of the study is to assess the suitability of two sorts of plastic residues: shredder light fraction and pellets made of selected plastic waste.

## 2. Dual fluidized bed gasification

At the Vienna University of Technology the dual fluidized bed gasification system has been developed in order to generate high quality producer gas. The dual fluidized bed gasifier is an allothermal gasifier, where gasification and combustion take place in spatially separated reactors. Both reactors are thermally connected due to a circulating bed material. Two different gas streams are yielded: producer gas with high calorific value and conventional flue gas. The principle is shown in Figure 1. In the gasification reactor, gasification takes place in a bubbling bed, which is fluidized with steam. Some ungasified char remains that is transported to the combustion reactor together with the circulating bed material. This char is combusted with air in a highly expanded fast fluidized bed. Heat is delivered back to the gasifier by the bed material to satisfy the endothermic gasification reactions.

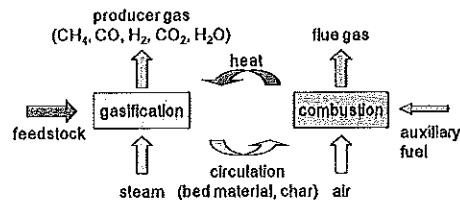


Fig. 1: Basic principle of the dual fluidized bed gasifier

This gasification process has been demonstrated successfully in Güssing (Austria). Producer gas is converted into electrical power in a gas engine. Heat generated in the process is fed into the local district heating system [6]. This technology is now commercially available.

## The 100 kW pilot plant

At Vienna University of Technology gasification tests are carried out in a 100 kW pilot plant. It is a dual fluidized bed gasifier similar to the demonstration plant in Güssing but smaller in size. In Figure 2 the pilot plant is schematically illustrated.

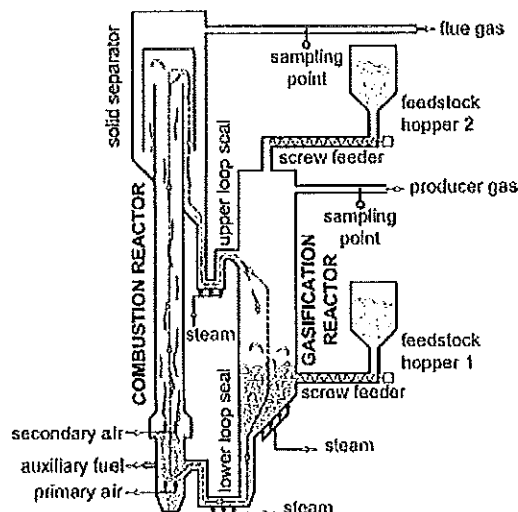


Fig. 2: 100 kW dual fluidized bed pilot plant

Feedstock is stored in different gas tight hoppers and transported with a screw feeding system directly into the fluidized bed. Plastic material is thrown onto the fluidized bed from the top of the gasifier.

This hopper has a water-cooled jacket in order to prevent melting of the plastic residues. In this gasification process olivine is used as bed material. Olivine shows moderate tar cracking activity and has good mechanical stability [7]. The gasification reactor is fluidized with superheated steam. Typical gasification temperatures amount to 800-850°C. The gasification and the combustion reactor are connected by loop seals. In order to promote transport of solids and to prevent gas leakage the loop seals are also fluidized with steam. In the combustion reactor heat is generated as residual char is combusted with air. At the bottom of the combustor primary air is injected and a dense fluidized bed is formed. Light fuel oil is used as auxiliary fuel there. The temperature in the gasifier is controlled by the amount of oil combusted. Secondary air is injected at a higher level to transport particles to the top of the riser. Hot bed material is precipitated from the flue gas stream and transported back into the gasification reactor.

Further facilities downstream the producer and flue gas line are not shown in Figure 2. Prior to gas sampling and analysis the producer gas is cooled in a heat exchanger to lower temperatures. After separated gas lines for producer gas and flue gas, both gas streams are mixed and combusted in an afterburning chamber with air. A cyclone removes particles prior to the stack.

### Measurement equipment

During the experiments, producer gas and flue gas properties are measured online. The main producer gas compounds are analyzed with a Rosemount NGA2000 device. A gas chromatograph (Syntech Spectras GC 955) is employed for the online-measurement of  $N_2$ ,  $C_2H_4$ ,  $C_2H_6$  and  $C_3H_8$ . An impinger bottle method for

tar measurement has been developed at Vienna University of Technology. It is similar to the conventional tar protocol, but has been adapted for producer gas from steam gasification. Toluene is used as a tar absorbent. Dust, entrained char, water and tar content can be analyzed from one sample. Two different types of tars are measured: Gravimetric tars are weighed after evaporation of the solvent. A GCMS device is used to measure the content of many different tar species. Further description of the tar measurement is available in [8].  $NH_3$ ,  $HCl$  and  $H_2S$  are also measured by an impinger bottle method. The ammonia content is determined by dissolution in  $H_2SO_4$ . Ammonium sulfate is formed and detected by ion chromatography. The concentration of  $HCl$  is analyzed by dissolution in  $H_2O_2$  in impinger bottles. Chlorine is measured by ion chromatography. For the measurement of  $H_2S$  impinger bottles filled with 35%- $KOH$  are used.  $H_2S$  is detected by potentiometric titration.

In the flue gas stream the  $CO$ ,  $CO_2$  and  $O_2$  content is measured by a Rosemount NGA2000 device. For the online measurement of  $NO$  a Rosemount NGA2000 MLT4 device is used, for  $SO_2$  it is a Binos 1004. The  $HCl$  concentration in the flue gas stream is analyzed in the same way as in the producer gas.

### 3. Feedstock characterization

Two different types of plastic residues are tested: plastics from shredder light fraction and pellets made of selected plastic waste. Plastics from shredder light fraction (SLF) is produced in a mechanical sorting plant, where different plastic residues (shredder light fraction, plastics and films from commercial waste and waste of electrical equipment) are processed. Shredder light fraction is

produced, when end-of-life vehicles are recycled, shredded and graded. Plastic pellets are produced from a mixture of classified municipal solid waste (MSW), plastics from bio-mechanical treatment of waste and plastic packaging from selective collection.

Both plastic residues contain a variety of different polymers and cannot be used in a material-sensitive way. An interesting application is the use as reducing agent in blast furnaces for steel production [9]. Most commonly, waste plastics are recovered thermally in waste incineration plants. Plastics from bio-mechanical treatment of waste are incinerated primarily in fluidized bed incinerators [10]. Automotive shredder residues and other plastic waste are also used as feedstock for a pyrolysis plant coupled to the boiler of a hard coal power plant [11]. Pyrolysis and combustion are thermal treatment processes, which are already used for plastic residues. Gasification is also an interesting approach and thus, experiments with plastic residues are conducted. Table 1 lists the proximate and the elementary analyses of the plastic feedstock used in the gasification experiments.

In order to point out differences plastic residues are compared to the standard fuel of the pilot plant, soft wood pellets (wood chips in industrial gasifiers). The lower calorific value (LCV) of both plastic residues is higher than soft wood. It is mainly influenced by the oxygen content of the fuel that is significantly lower in plastic residues. The concentration of elements is given on a water and ash free basis (waf). The carbon content of plastic residues is higher, which can be explained by the molecular structure of polymers that is rich of carbon for the majority of polymers.

	SLR-plastics	MSW-plastics	soft wood
LCV, kJ/kg	31946	24092	17458
water, wt.-%	0.87	2.81	6.11
ash, wt.-%, dry	10.67	12.47	0.29
C, waf	72.76	61.88	50.38
H, waf	8.90	8.36	6.06
O, waf	15.08	27.51	43.51
N, waf	1.04	1.07	0.05
S, waf	0.35	0.24	0.005
Cl, waf	1.87	0.94	0.003
volatiles, waf	89.2	90.2	86.7

Tab. 1: Feedstock analysis of plastic residues and soft wood

In particular, the concentrations of compounds that form undesired pollutants during gasification are of interest. Nitrogen, sulfur and chlorine are trace compounds in wood, but are present in plastic residues in the range of 1%. The content of volatiles allows a first assessment of the devolatilization and gasification behavior of the fuels. More volatile matter and less char are found in plastic residues. It indicates that more auxiliary fuel is necessary for gasification, because there is less char available for combustion. The ash content of both plastic residues is considerably high owing to the origin of the fuels.

	SLR-plastics	MSW-plastics	wood
DT, °C	1200	1170	1400
FT, °C	1320	1230	1450

Tab. 2: Deformation (DT) and flow temperatures (FT)

The ash melting behavior is crucial for the operation of fluidized beds. Table 2 gives the deformation temperature (DT) and the flow temperature (FT) of all samples used during this investigation. The temperatures, which occur in the combustion reactor, are in the range of 890°C to 920°C. As the deformation temperature of all ash samples is considerably higher, no problems related to ash melting are expected.

#### 4. Results and discussion

Several test series are performed in order to assess the particular gasification behavior of plastic residues. They are presented in Table 3.

In series I, both plastic residues are mixed with wood pellets. The experiments are conducted each with 50% fuel power from plastic residues and 50% from wood pellets. In series II, the share of SLF-plastics is increased from 0% (pure wood) to 25%, 50%, 75% and 100%. Variation of steam/carbon ratio is the focus of series III. Two different mixtures of SLF-plastics, 25% and 50%, and one mixture of MSW-plastics are gasified with varying amounts of steam.

#### Operation parameters

During all experiments, the main operation parameters are kept constant, so that the results are comparable. The thermal fuel power is set to 100 kW ( $\pm 3$  kW) and the gasification temperature is kept constant at 850°C ( $\pm 4$ °C) in all experiments. The temperature is measured in the fluidized bed of the gasification reactor. The steam/carbon ratio (S/C) is defined as the ratio of the sum of the amount of steam used for fluidization and the moisture content of the feedstock (kg/h) to the amount of carbon in the feedstock (kg/h). It is kept constant at 1.80 kg/kg ( $\pm 0.06$  kg/kg) during all experiments if possible. When 100% SLF-plastics are gasified, the total fuel input is 89 kW. Due to limitations of the feeding system, it is not possible to further increase the mass flow. Concerning fuel load, the experiments are nevertheless comparable according to experiences with other fuels. Moreover, the S/C ratio is raised to 2.0 kg/kg. In order to maintain the circulation and the sufficient fluidization of the fluidized bed, a minimum amount of steam is required, which cannot be further reduced. The influence of variation of S/C is discussed in series III.

	fuel mixture			S/C				
	SLF-plastics	MSW-plastics	wood	1.4	1.6	1.8	2.0	
series I	0%	0%	100%			x		series III
	50%	0%	50%		x	x		
	0%	50%	50%		x	x		
series II	25%	0%	75%	x		x		
	75%	0%	25%			x		
	100%	0%	0%				x	

Tab. 3: Overview on experimental series

## Auxiliary fuel demand

In the dual fluidized bed gasification system, heat is consumed in the gasification reactor because of the endothermic gasification reactions. In the combustion reactor, heat is provided due to combustion of residual char from gasification and auxiliary fuel. If no auxiliary fuel is added, the temperature in the gasification reactor is leveled according to the energy demand of the gasification reactions and the amount of char transported into the combustion reactor together with the bed material. In order to control the gasification temperature and to keep it at 850°C, auxiliary fuel is necessary for wood as well as for plastics. In the pilot plant light fuel oil is used as auxiliary fuel.

	aux. fuel kg/h	fuel input kW
50% SLF	3.2	98
50% MSW	3.0	99
100% wood	2.9	97
100% wood	2.9	97
50% SLF	3.2	98
100% SLF	3.2	89

Tab. 4: Auxiliary fuel input to the combustor and fuel input to the gasifier

It is found that more auxiliary fuel is required for gasification of plastic residues compared to wood pellets; data is compiled in Table 4. The main reason is that less char is available after devolatilization of plastic residues. In order to ensure constant gasification temperature, more fuel oil has to be inserted into the combustor. In series II it is observed that the auxiliary fuel demand in the combustor increases with increasing share of SLF-plastics. When only SLF-plastics are gasified, 3.2 kg/h of light fuel oil are necessary. As the fuel

power of the gasifier was reduced, the demand of auxiliary fuel would be higher at 100 kW (3.5 kg/h).

## Main producer gas compounds

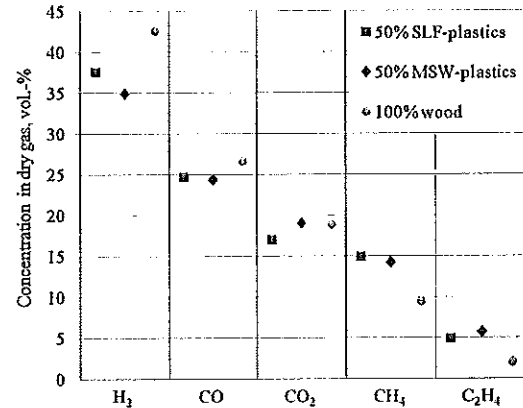


Fig. 3: Main producer gas compounds (series I)

Figure 3 presents the average producer gas composition during gasification of mixtures of plastic residues and wood (series I). Both plastic residues increase the yield of methane, C<sub>2</sub>H<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, and C<sub>3</sub>H<sub>8</sub> compared to wood (Figure 4). Those compounds are decomposition products of polymers. The concentration of H<sub>2</sub> decreases, as the concentration of other compounds, which contain hydrogen, increase (CH<sub>4</sub>, C<sub>2</sub>H<sub>4</sub>). The oxygen content of the feedstock is lower. Therefore, the producer gas compounds that contain oxygen (CO and CO<sub>2</sub>) decrease. The LCV of the dry producer gas from the two plastic residues amounts to 16 MJ/m<sup>3</sup> (stp = standard temperature and pressure of 273.15K and 101325 Pa).

The majority of nitrogen and sulfur in the fuel is converted into NH<sub>3</sub> and H<sub>2</sub>S in the producer gas. HCl is mainly bound to ash particles [12]. Gasification of polluted feedstock increases the concentration of pollutants in the producer gas, which depicts Figure 4. The concentration of HCl in the producer gas is higher, when MSW-plastics are gasified, although the concentration of chlorine in MSW-

plastics is lower than in SLF-plastics. This indicates that there is more chlorine in the char from SLF-plastics, which is transported to the combustion reactor. More details on that are available in the section on flue gas.

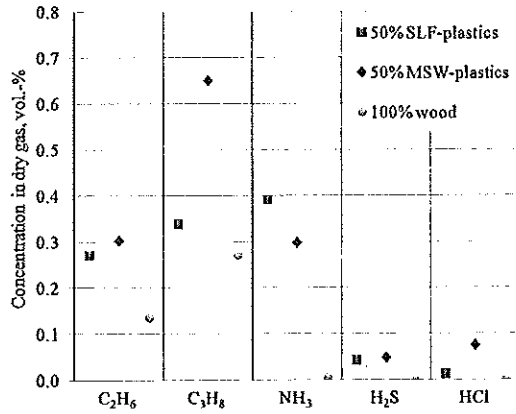


Fig. 4: Minor producer gas compounds (series I)

The change in gas composition with increasing share of SLF-plastics is investigated in series II. It is found that the change is non-linear (Figure 5). When a mixture of wood and plastic residues is gasified, the decomposition products of the different feedstock react with each other, which results in different gas compositions. As there is less oxygen in SLF-plastics, the concentration of CO lessens markedly. A slight decrease of CO<sub>2</sub> is also observed. The rise of CH<sub>4</sub> is almost linear. 100% of SLF-plastics yield more than double of the concentration of methane compared to wood; the LCV of the dry producer gas amounts to 17.8 MJ/m<sup>3</sup> (stp). The H<sub>2</sub> content is first decreasing and with higher shares of plastics again increasing. This is most probably due to two different mechanisms. At high shares of wood and low shares of plastics oxygen is necessary for gasification of char from wood to produce CO and additional H<sub>2</sub> is set free from steam. With higher shares of plastics and lower shares of wood the amount of char decreases and, therefore, less steam is necessary for char gasification. But

then another fact becomes more important. Plastics contain less oxygen (Table 1) and for equilibrium reasons oxygen is necessary for production of CO and CO<sub>2</sub>. Therefore, with increase in the share of plastics more steam reacts with hydrocarbons and again more additional H<sub>2</sub> is set free into the producer gas.

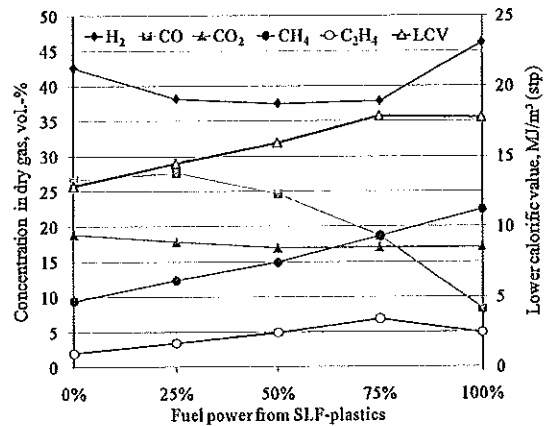


Fig. 5: Change in gas composition with increasing share of SLF-plastics, 0% = pure wood

During series III the steam/carbon ratio is varied. Figure 6 shows the change in concentration of producer gas compounds in percentage points, if the S/C ratio is lowered. The amount of steam is reduced to lower the S/C ratio.

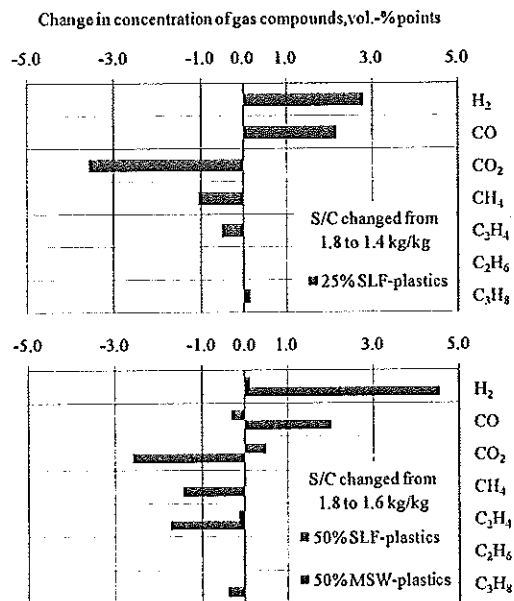


Fig. 6: Change in gas composition with changing S/C ratio (series III)

In the upper part of Figure 6, the S/C ratio is changed from 1.8 to 1.4 kg/kg; in the lower part the change from 1.8 to 1.6 kg/kg is illustrated. Gasification of 25% SLF-plastics and 50% MSW-plastics show the same tendency: H<sub>2</sub> and CO increase, all other compounds decrease. When pure wood is gasified, H<sub>2</sub> and CO<sub>2</sub> decreases and CO and CH<sub>4</sub> content increases with decrease in S/C. Further information on that is available in [13]. Apparently, gasification of wood is more influenced by changes in steam mass flow. The reaction of solid carbon with steam is likely to be highly influenced thereby. When 50% SLF-plastics are gasified, the gas composition remains almost constant, when S/C is lowered. As there is more char in wood, more significant changes occur, when 25% SLF-plastics and 75% wood are gasified.

### Tars, dust, and char

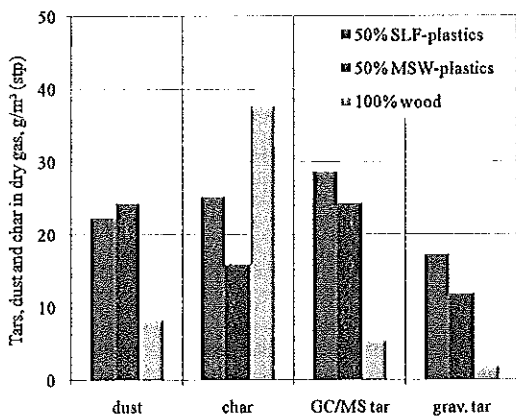


Fig. 7: Tars, dust, and char in producer gas (series I)

Figure 7 depicts the solids concentration in the producer gas stream. Due to the high ash content of plastic residues, there is more dust in the producer gas. As a consequence of the lower amount of char in plastic residues, less char is entrained by the gas flow compared to wood gasification. Gasification of plastic residues increases the amount of gravimetric tar and GC/MS tar

considerably. Decomposition of polymers does not only produce CH<sub>4</sub>, C<sub>2</sub>H<sub>4</sub>, etc, but also higher hydrocarbons.

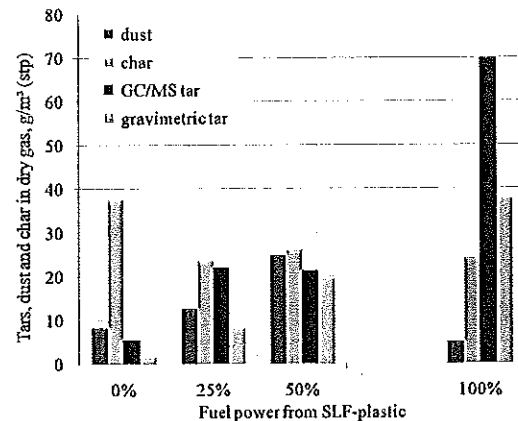


Fig. 8: Change in concentration with increasing share of SLF-plastics

Series II confirms the findings described above, the results are compiled in Figure 8. With increase in the share of SLF-plastics, the concentration of gravimetric and GC/MS tar increases markedly. The reduction of dust and char is caused by the lower fuel input to the gasifier (89 kW) and therefore the lower mass flow of SLF-plastic.

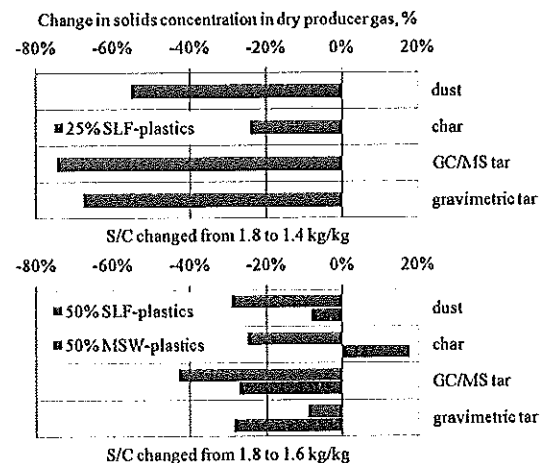


Fig. 9: Change in concentration with changing S/C ratio

Figure 9 shows the influence of S/C on tar formation (upper part: change from 1.8 to 1.4 kg/kg, lower part: from 1.8 to 1.6 kg/kg). It has to be emphasized that



wood is fed directly into the fluidized bed, and both plastic residues are thrown onto the fluidized bed. With increase in steam mass flow, the gas volume and the velocity increases and more unreacted particles that form tars, are carried out of the reactor. Hence, lower S/C ratios lead to lower solids concentration in the producer gas. The only exception is the increase in char with decreasing S/C for 50% of MSW-plastics. There is no plausible explanation for that observation yet.

### Flue gas composition

		50% SLF	50% MSW
NO	mg/m <sup>3</sup> (stp)	60	60
SO <sub>2</sub>	mg/m <sup>3</sup> (stp)	<1	<1
HCl	mg/m <sup>3</sup> (stp)	134	32

Tab. 5: Pollutants in flue gas (series I)

When mixtures of 50% wood and 50% plastic residues are gasified during series I, the flue gas composition is also investigated. The flue gas from the combustion reactor is conventional flue gas, consisting of approx. 12% CO<sub>2</sub>, 5% O<sub>2</sub>, and 83% N<sub>2</sub>. The concentrations of NO, SO<sub>2</sub> and HCl, referred to 6% O<sub>2</sub> in the flue gas, are compiled in Table 5. The concentration of HCl is quite high and has to be considered in the design of the flue gas cleaning equipment. More HCl is found in the flue gas of SLF-plastics, in turn, the HCl content in the producer gas is lower than expected. Apparently, there is more chlorine in the char of SLF-plastics.

### 5. Comparison to wood pellets

In order to highlight differences in gasification behavior, gasification of

100% SLF-plastics is compared to gasification of 100% soft wood pellets.

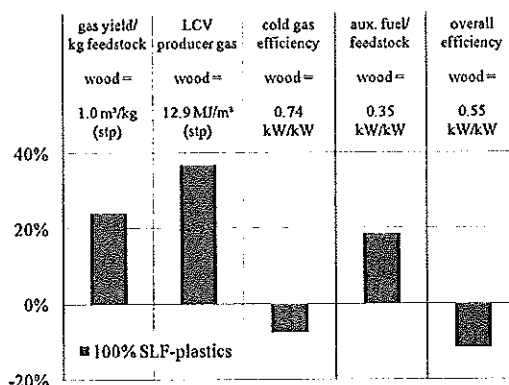


Fig. 10: Deviation of gasification of SLF-plastics from wood pellets

In Figure 10 the deviation from gasification of wood pellets as base case is illustrated. Producer gas yield in relation to the mass flow of feedstock increases, when plastics residues are gasified. This is due to the higher lower calorific value (LCV) of the material, but most notably due to increasing reaction with the steam. More steam is converted into producer gas compounds during gasification of plastics. As mentioned before, plastics lead to higher LCV of the producer gas than wood. Cold gas efficiency is defined as the ratio of chemical energy in the producer gas and chemical energy of the feedstock. The ratio of auxiliary fuel demand and energy content of the feedstock is 0.35 kW/kW in case of wood gasification. This value depends on the char content of the feedstock; hence plastics require more fuel oil. The overall efficiency also takes into account the auxiliary fuel demand. Associated to the high amount of auxiliary fuel, lower overall efficiency is reached for gasification of plastics. As significant thermal losses occur in the pilot plant due to the disadvantageous ratio of volume and surface and the quality of insulation, the values for cold gas efficiencies from the pilot plant cannot be applied directly to industrial

scale units. In industrial size units, where specific heat losses are smaller and feed stream preheating is included, the cold gas efficiency and especially the overall efficiency are higher than the values for the pilot plant displayed in Figure 10.

## 6. Conclusion

Two different types of plastic residues are gasified in a dual fluidized bed gasifier. In order to investigate the key parameters of gasification, different mixtures with soft wood pellets ranging from 0-100%, as well as different steam-to-carbon ratios are tested. It is found that increasing share of plastics in the fuel mixture leads to higher concentrations of CH<sub>4</sub>, C<sub>2</sub>H<sub>4</sub> and other light hydrocarbons in the producer gas. The tar content of the producer gas increases, whereas entrained dust and char decreases. Mixtures of plastics and wood change the producer gas composition in a non-linear way, which indicates alternately influence of both feedstock. An increasing demand of auxiliary fuel is observed, which originates from the lower char content in plastic residues.

## 7. Acknowledgements

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