# Steam gasification of sewage sludge for synthesis processes

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**Abstract**

The paper presents measurement results of a gasification test run. Municipal sewage sludge from a digestion tower is gasified in an advanced dual fluidized bed reactor system. Steam is used as gasification agent and an olivine-limestone mixture as bed material. The fuel analysis shows a very high ash content and a low heating value of the dried sewage sludge. In addition, a significant amount of nitrogen in the fuel is present, leading to a high ammonia content in the product gas. Sintering effects caused by the high ash content do not occur. Thus, a gasification process without limitation is achieved. The fuel input is located in the lower gasification reactor operating as bubbling fluidized bed, whereas the upper gasification reactor is designed as a column of turbulent fluidized zones for tar cracking. The results show an efficient in-situ tar reduction. With a look on the product gas composition a comparatively high carbon dioxide and a low carbon monoxide content is surprising. It is obvious that an iron oxide reduction of the initial fuel ash occur in the gasification reactor. In addition, it is assumed that the significant iron content in the fuel ash also leads to a transport of oxygen from the combustion reactor to the gasification reactor. Thus, carbon monoxide and hydrogen are oxidized in the gasification reactor by the circulating iron-rich ash particles (chemical looping effect).

## 1. Introduction

The thermochemical conversion of biomass is a promising option for the environmentally friendly production of valuable renewable products. Fuels from biogenic sources are particularly relevant, as these fuels constitute the only practical carbon source available within the range of renewables. Fluidized bed steam gasification of solid fuels is a promising technology to produce a nearly nitrogen-free and hydrogen-rich product gas. After gas cleaning processes, the utilization of the product gas as syngas for various synthesis processes is auspicious. [1]

In the case of industrial utilization, high quality wood chips are a comparatively expensive fuel [2]. Waste material and biogenic residues as fuel, like sewage sludge, are of interest for gasification processes and for the future energy supply. However, alternative low-cost fuels often have difficult chemical and physical properties. A risk that higher hydrocarbons such as tar will not convert sufficiently is present. In addition, undesired substances, high contents of fuel ash and disadvantageous ash sintering behavior lead to limitations regarding fluidized bed gasification. This may affect the plant availability in a critical way. [3]

To face these challenges, an advanced gasification reactor design was developed and a pilot plant with 100 kW fuel power was put in operation at TU Wien in 2014. Fig. 1 shows the basic idea of the advanced gasification process. The special design of the gasification reactor aims at supporting gasification reactions, improving conversion rates, and minimizing the tar content of the product gas. Fig. 2 shows an illustration of the pilot plant at TU Wien. The upper part of the gasification reactor features a column of fluidized bed zones with counter-current flow of the raw product gas flowing upwards and hot bed material introduced from the combustion reactor flowing downwards. Thus, the increased contact between the introduced fuel, the volatiles, and the bed material provides for a significant reduction of undesired product gas components like tar [4,5]. The 100 kW fuel power pilot plant enables gasification test runs with a relevant plant size. This is indispensable to gather meaningful results.

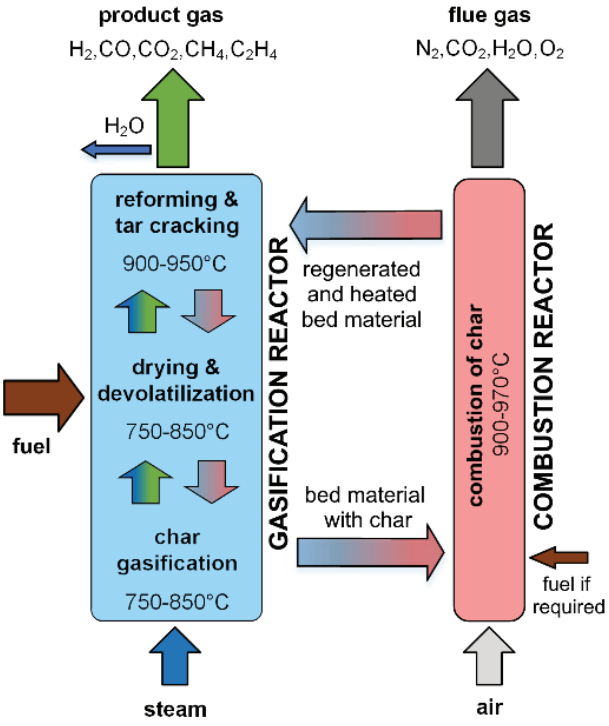


**Fig.1: Basic principle of dual fluidized bed steam gasification at TU Wien**

## 2. Methodology

The investigations base on experimental research with the advanced 100 kW pilot plant at TU Wien. Fig. 2 shows the inner dimensions and heights of the used dual fluidized bed gasifier. All pressure and temperature measurement points of the reactor system are visible. A screw conveyor feeds the solid fuel onto the bubbling bed of the lower gasification reactor. A special feeding system enables the utilization of a broad range of fuels without limitations.

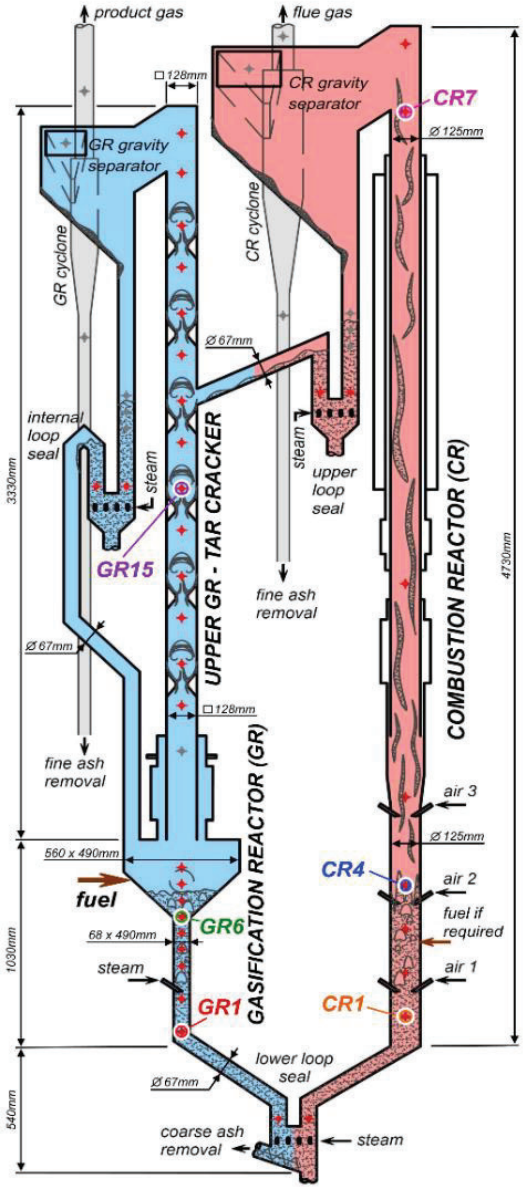


**Fig.2: Dimensions, media inputs and measurement points of the gasifier**

The pilot plant is equipped with a programmable logic controller (PLC), which continuously gathers and records all measured data. The installation of more than 20 media flows, 100 temperatures, and 70 pressures ensure an insight into the gasification process. Different Rosemount NGA2000 gas analyzers continuously measure the main product gas and flue gas components like hydrogen ($H_2$), carbon monoxide (CO), carbon dioxide ($CO_2$), methane ($CH_4$) and oxygen ($O_2$). Perkin Elmer ARNEL – Clarus 500 gas chromatograph measures further product gas components, like ethylene ($C_2H_4$) and ethane ($C_2H_6$) every 15 minutes. The setup of the product gas and flue gas measurement can be seen in Fig. 3. The product gas has to be filtered and washed with rapeseed methyl ester (RME) in advance to eliminate fine dust particles and condensable components like water and tar.

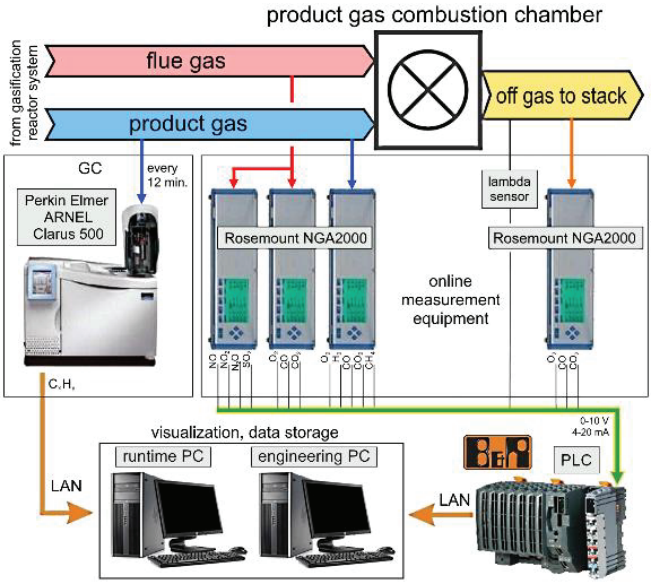


**Fig.3: Online measurement of gases [4]**

Other product gas components like tar, water, char and dust are detected discontinuously (Fig. 4). Solid particles, like char and dust are sampled with a small cyclone and a quartz wool stuffed filter cartridge. Water and tar is sampled with impinger bottles and toluene as solvent.

Following, the gravimetric tar content as well as tar components detected by gas chromatography coupled with mass spectrometry (GCMS) are determined. The measurement method is based on CEN/TS 15439. The sampling procedure for the detection of ammonia ($NH_3$) and hydrogen sulfide ($H_2S$) in the product gas is very similar, but with the use of different solvents.

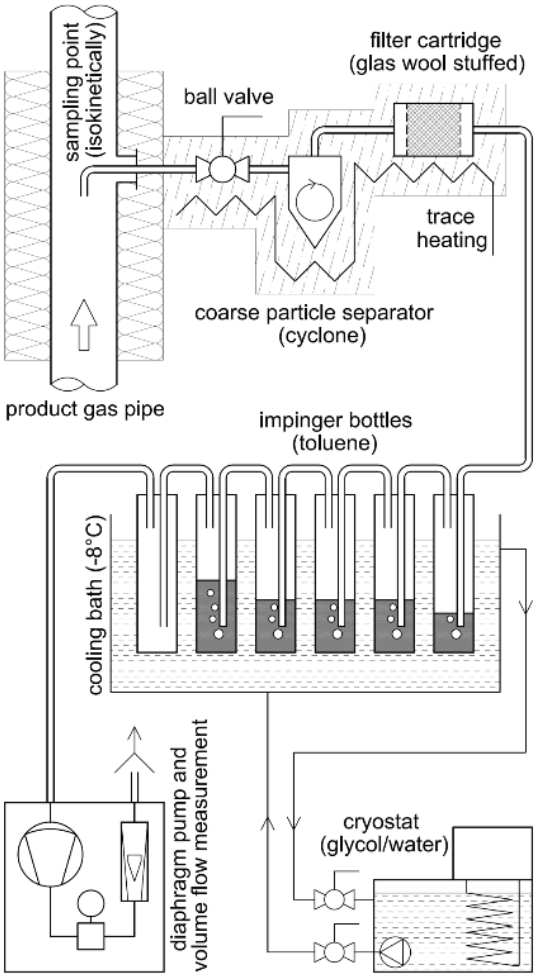


**Fig.4: Tar, dust, char and water sampling [6]**

This paper presents the measurement results of a steam gasification test run with municipal sewage sludge from digestion towers as fuel and a mixture of olivine and limestone as bed material. The extensive scientific equipment gathers measurement values of the experiment. A detailed description of the pilot plant at TU Wien is available in literature [6,7,8].

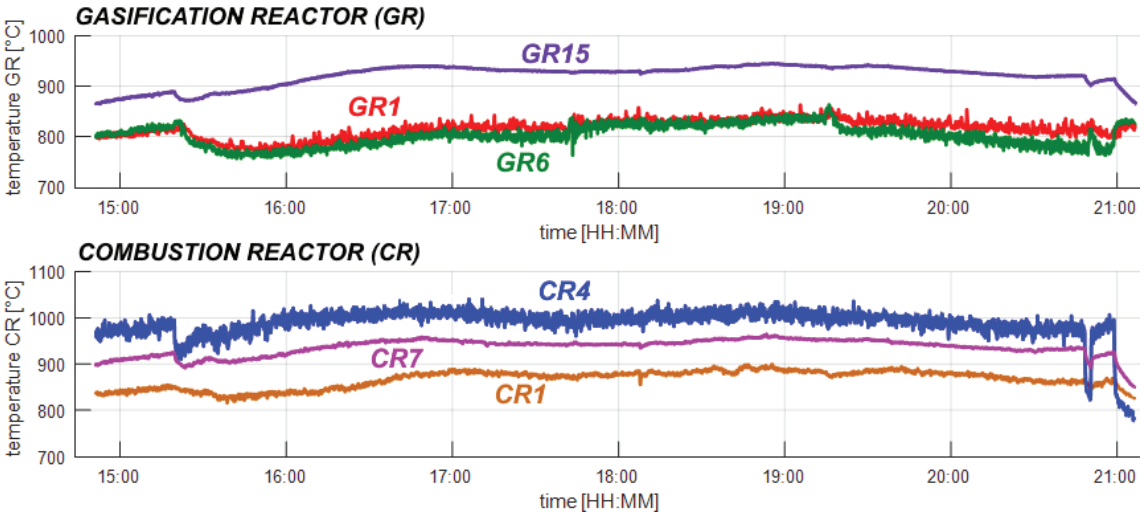


**Fig.5: Courses of specific temperatures during the gasification test run over time**

This paper focuses on the visualization of pure measurement data visualized with Tables and Figures. For the evaluation of measurements, typically the mean values of a defined steady-state operation phase are used. Fig. 5 shows the courses of temperatures over time of the test run. It is shown that a uniform operation was achieved. For the heat-up procedure of the pilot plant softwood was used. The gasification of 100 % sewage sludge pellets took place between 15:30-17:40 and 19:20-20:45. Softwood pellets were gasified in between due to limited amount of pelletized sewage sludge and the preparation work for measurement procedures of ammonia ($NH_3$) and hydrogen sulfide ($H_2S$). Fig. 6 shows the temperature profiles along the reactor height for the steady-state phase 17:10-17:40. A simulation with the software tool IPSEpro is in progress. It will enable calculations of mass and energy balances and will give a deep insight into the test run in future.

## 3. Results and Discussion

Table 1 presents main process parameters of the gasification test run for two defined steady-state operation phases. Table 2 shows the fuel analysis of sewage sludge. The fuel was dried and pelletized to guarantee a constant fuel feeding into the gasification reactor via a screw conveyor. The high ash content causes a low heating value. A high amount of nitrogen is present in sewage sludge as fuel.

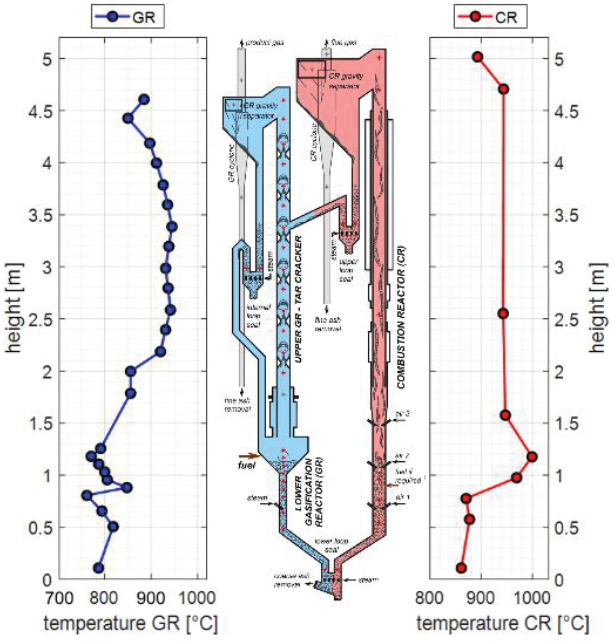


**Fig.6: Temperature profiles of the gasification and combustion reactor (mean 17:10-17:40)**

High ash melting temperatures predict an operation without sintering effects of the bed material particles caused by the fuel ash. In addition, Table 3 shows the XRF-analysis of the fuel ash. Attention should be paid on the relatively high content of iron oxide as an integral part of the ash. Iron (iron oxide) is a suitable oxygen carrier in dual fluidized beds at high temperatures [9].

**Table 1: General process parameters of the sewage sludge gasification test run**

| parameter | unit | mean value | |
|---|---|---|---|
| **bed material** | wt.-% | 80% olivine, 20% limestone | |
| **steam/fuel ratio** | $kg_{H2O}/kg_{fuel,daf}$ | 1.45 | |
| **steam/carbon ratio** | $kg_{H2O}/kg_{C}$ | 2.72 | |
| **fuel input GR** | kW (kg/h) | 69.8 (28.1) | |
| **steady-state phase** | hh:mm | 16:10-16:40 | 17:10-17:40 |
| **additional fuel input CR** | kW | 76.3 | 51.7 |
| **temp. in lower GR (GR6-GR1)** | °C | 780-800 | 800-815 |
| **temp. in upper GR (GR15)** | °C | 925-935 | 935-940 |
| **temp. in CR (CR7)** | °C | 930-940 | 940-950 |

**Table 2: Fuel analysis of municipal sewage sludge from a digestion tower from Krems/Austria**

| parameter | unit | value |
|---|---|---|
| **water content** | wt.-% | 8.7 |
| **ash content** | $wt.-\%_{db}$ | 52.3 |
| **carbon (C)** | $wt.-\%_{db}$ | 25.48 |
| **hydrogen (H)** | $wt.-\%_{db}$ | 3.02 |
| **nitrogen (N)** | $wt.-\%_{db}$ | 3.46 |
| **sulfur (S)** | $wt.-\%_{db}$ | 1.18 |
| **chlorine (Cl)** | $wt.-\%_{db}$ | 0.106 |
| **oxygen (O)*** | $wt.-\%_{db}$ | 14.45 |
| **volatile matter** | $wt.-\%_{db}$ | 44.55 |
| **lower heating value, moist** | MJ/kg | 8.94 |
| **ash deformation temp. (A)** | °C | 1140 |
| **ash flow temp. (D)** | °C | 1230 |

* calculated by difference to 100 $wt.-\%_{db}$

Fig. 7 shows a section of the main product gas components of the experimental test run over time. The mean values of the product gas composition and the content of different minor components in the product gas are presented in Table 4. The results present the product gas composition without nitrogen from purging the fuel hoppers and the measurement points. Therefore, only the nitrogen in the fuel causes the presented nitrogen ($N_2$) and ammonia ($NH_3$) content in the product gas. It is assumed that the not measured hydrogen cyanide (HCN) is a product gas component, approximately ca. 8-9 % of the ammonia value [10].

**Table 3: XRF-analysis of sewage sludge ash**

| oxide | content wt.-% | oxide | content wt.-% |
|---|---|---|---|
| **$MoO_3$** | n.d. | **$Cr_2O_3$** | 0.04 |
| **$Nb_2O_5$** | n.d. | **$V_2O_5$** | n.d. |
| **$ZrO_2$** | 0.06 | **$TiO_2$** | 0.83 |
| **SrO** | 0.11 | **CaO** | 13.18 |
| **PbO** | 0.01 | **$K_2O$** | 1.29 |
| **$As_2O_3$** | 0.01 | **Cl** | 0.14 |
| **ZnO** | 0.29 | **$SO_3$** | 4.80 |
| **CuO** | 0.11 | **$P_2O_5$** | 14.85 |
| **NiO** | 0.02 | **$SiO_2$** | 33.23 |
| **$Co_3O_4$** | n.d. | **$Al_2O_3$** | 17.59 |
| **$Fe_2O_3$** | 8.50 | **MgO** | 3.52 |
| **MnO** | 0.07 | **$Na_2O$** | 1.37 |

n.d. not detected

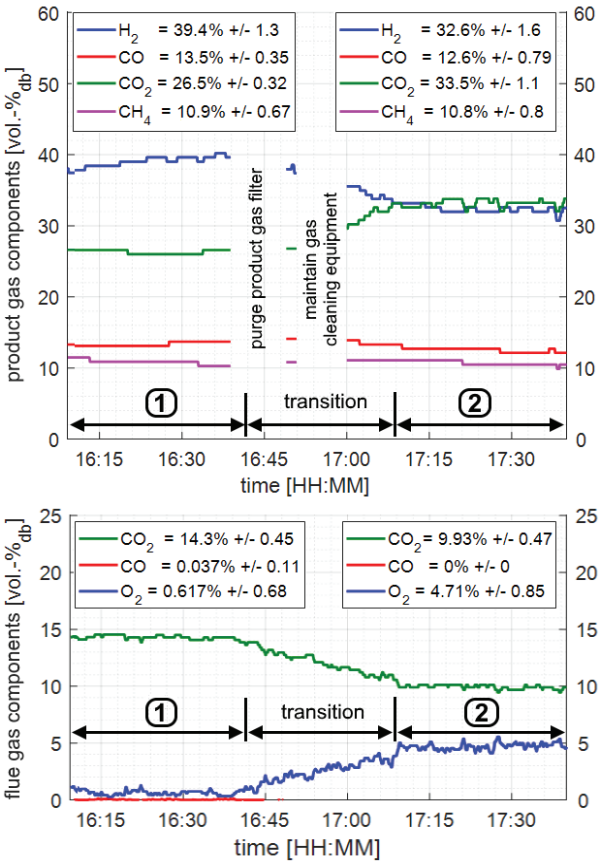


**Fig.7: Courses over time of product gas and flue gas components during the gasification test run**

**Table 4: Product gas components of sewage sludge steam gasification**

| parameter | unit | before upper GR (before tar cracker) | after upper GR (after tar cracker) | |
|---|---|---|---|---|
| | | | 16:10-16:40 | 17:10-17:40 |
| **hydrogen ($H_2$)** | vol.-%$_{db}$ | n.p. | 39.4 | 32.6 |
| **carbon monoxide (CO)** | vol.-%$_{db}$ | n.p. | 13.5 | 12.6 |
| **carbon dioxide ($CO_2$)** | vol.-%$_{db}$ | n.p. | 26.5 | 33.5 |
| **methane ($CH_4$)** | vol.-%$_{db}$ | n.p. | 10.9 | 10.8 |
| **ethylene ($C_2H_4$)** | vol.-%$_{db}$ | n.p. | 3.0 | 2.3 |
| **ethane ($C_2H_6$)** | vol.-%$_{db}$ | n.p. | 0.19 | 0.11 |
| **nitrogen ($N_2$)*** | vol.-%$_{db}$ | n.p. | 0.2-0.9 | 2.1-2.2 |
| **ammonia ($NH_3$)** | vol.-%$_{db}$ | n.p. | n.p. | 4.6 |
| **hydrogen sulfide ($H_2S$)** | vol.-%$_{db}$ | n.p. | n.p. | 1.4 |
| **fly char** | g/Nm$^3_{db}$ | 26.3 | n.p. | 1.1 |
| **dust** | g/Nm$^3_{db}$ | 1350 | n.p. | 8.1 |
| **water content** | vol.-% | 55 | n.p. | 62 |
| **benzene (B)** | mg/Nm$^3_{db}$ | 21650 | n.p. | 7300 |
| **toluene (T)** | mg/Nm$^3_{db}$ | n.m. | n.p. | n.m. |
| **ethyl-benzene (E)** | mg/Nm$^3_{db}$ | 840 | n.p. | 51 |
| **xylenes (X)** | mg/Nm$^3_{db}$ | 2340 | n.p. | 12 |
| **GCMS tar (without BTEX)** | g/Nm$^3_{db}$ | 27.5 | n.p. | 16.3 |
| **grav. tar** | g/Nm$^3_{db}$ | 35.5 | n.p. | 4.75 |

n.p.: measurement not performed, n.m.: not measureable (toluene is solvent for tar sampling procedure), * nitrogen from purging fuel hoppers and measurement points excluded

The results show that the special design of the gasification reactor is favorable to decrease undesired components in the product gas. Detectable tar contents in the product gas (GCMS, grav. tar, BTEX) decrease significantly during the flow through the upper gasification reactor. Especially, the reduction of the gravimetric tar by 87 % has to be mentioned, because gravimetric tar represents the problematic heavy polycyclic aromatic hydrocarbons in the product gas. Heavy polycyclic aromatic hydrocarbons are the main reason for fouling and clogging effects inside product gas pipes and apparatuses of gasification plants. Toluene is used as solvent for the tar sampling procedure. Thus, it is not possible to detect the toluene content in the product gas. Experience shows that typical toluene contents are in the range of 20-50 % of the benzene value.

Fig. 7 has to be discussed in detail. The measurement values show that a reduction of the iron oxide of the fuel ash is present in the gasification reactor. It is assumed that an oxygen transport from the combustion to the gasification reactor occurred, which has to be validated by the use of process simulation. Two different steady-state operation phases were

reached during the gasification test run with sewage sludge as fuel. Phase 1 represents an operation where oxygen, introduced into the combustion reactor with the air, is fully utilized for the combustion reactions of char from the gasifier and additional fuel. After a transition period the steady-state phase 2 is reached. Phase 2 represents an operation with lower additional fuel input and/or higher air input into the combustion reactor. In that point, oxygen is present in sufficient amounts for an efficient oxidation of fuel ash iron (iron oxide). The reducing atmosphere and the high temperature in the gasification reactor promote the discharge of oxygen from the fuel ash. The release of oxygen results in lower hydrogen ($H_2$) and carbon monoxide (CO) and higher carbon dioxide ($CO_2$) and water contents ($H_2O$) in the product gas. It is obvious that the transport of oxygen from the combustion reactor to the gasification reactor is significantly higher in phase 2 in comparison to phase 1. This so-called chemical looping process is limited by the availability of oxygen in the combustion reactor. For phase 2 the high availability of oxygen ($O_2$) is visible with a look on the flue gas composition of Fig. 7.

It has to be mentioned, that the advanced reactor concept was designed for the reduction of tar in gasification applications and the possibility to produced $CO_2$ with chemical looping applications [11, 12]. Thus, a chemical looping active ash or bed material have significant higher effect and influence in the pilot plant at TU Wien compared to other dual fluidized bed designs. Actual experimental results show that the advanced fluidized bed reactor concept is predestinated for chemical looping combustion of solid fuels, especially biogenic residues and ligno-cellulosic biomass [13].

It is possible to present pictures (Fig. 8) of samples of the “oxidized” ash and bed material mix from the combustion reactor entering the gasification reactor, and the “reduced” ash and bed material mix leaving the gasification reactor in the lower part of the reactor system. The samples were taken after 4 hours of operation. The accumulation of bed ash from the sewage sludge in the bed material circulation is visible. Fig. 8 shows that a significant difference of the oxidation state of the iron content is present. The difference of the brown-red colored bed material entering the gasification reactor and the grey colored bed material leaving the gasification reactor is clearly visible. The presented results seems to confirm the theoretical discussion and the assumption of a possible chemical looping effect in dual fluidized bed gasifier systems if sewage sludge is utilized as fuel [14].

**Fig.8: Bed material ash mixture from the combustion reactor entering the gasification reactor (left: brown-red colored) and leaving the gasification reactor (right: gray colored)**

Additionally, also continuously introduced calcium carbonate from the fuel ash has to be considered as possible source of carbon dioxide in the product gas. Calcium carbonate ($CaCO_3$) forms calcium oxide (CaO) and releases carbon dioxide ($CO_2$) at high temperatures in the gasification reactor.

## 4. Conclusion and Outlook

Economic reasons lead to a rising interest to use alternative low-cost fuels for gasification processes. At the same time, products that are more valuable should be generated via gas upgrading or syntheses instead of heat and power generation. Thus, the focus is the production of hydrogen–methane gas mixtures [15], pure hydrogen [16], synthetic natural gas (SNG) [3,17,18], Fischer-Tropsch liquids [19] or mixed alcohols [20]. In addition, the direct utilization of the product gas as fuel gas is promising, directly substituting fossil fuels for high-temperature processes in industry [2,17].

In Austria approx. 236 000 t municipal sewage sludge (dry substance) were produced in 2017 from municipal sewage treatment plants with a capacity of at least 2000 PE60 (Population equivalent with a biochemical oxygen demand of 60g/day and person). 20% is introduced onto agricultural areas, 55% are treated thermally, and 25% are treated in another way (e.g. composting, mechanical-biological treatment) [21]. Therefore, sewage sludge has the utilization potential as fuel for dual fluidized bed steam gasification plants with a cumulative fuel power of 92 MW (7000 hours of operation per year). The 55% of sewage sludge already used in thermal processes would lead to 50 MW. Furthermore, sewage sludge is a promising fuel since currently municipal sewage treatment plants have to pay for disposal.

The measurement results of the pilot plant at TU Wien enable the generation of meaningful knowledge. This is especially relevant since the gasification of alternative and ash-rich fuel types is technologically challenging. The novel reactor design shows advantages to produce a product gas with comparable low tar contents. A chemical looping effect of the iron rich fuel ash could be responsible for the high carbon dioxide content in the product gas. The more oxygen is available in the combustion reactor, the higher is the transport of oxygen with the fuel ash from the combustion reactor to the gasification reactor. The product gas composition is influenced significantly. Nevertheless, a constant operation without problems was achieved with sewage sludge as fuel for a total period of nearly 4 hours. Important plant equipment like coarse ash output and fine/fly ash cyclones allowed a steady-state operation. Therefore, measures such as ash sieving procedures, hot bed material reuse, fuel blends variations, and varying bed material mixtures should be considered in future. To improve gasification efficiency or increase the hydrogen content in the product gas sorption enhanced gasification/reforming [4, 22] or a general operation with lower temperatures are promising options.

The pursuing research deals with detailed calculations regarding the presented sewage sludge test run. Performance indicating key figures have to be supported by the detailed validation of measurement values via mass and energy balance. The use of the simulation software IPSEpro enables the calculation of these figures. In addition, the presence of the chemical looping effect can be mathematically verified. Thus, the gained results and future calculations will generate knowledge for the up-scaling and design of industrial-sized dual fluidized bed gasifiers.

## 5. Acknowledgement

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