

Circulating Fluidized Bed Gasification of Solid Recovered Fuels - Results from Gasification and Gas Filtration Tests

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Fluidized bed gasification technology offers an attractive way to use inhomogeneous biomass residues and various waste-derived feedstocks in power and heat production as well as in various circular economy concepts based on the synthesis gas route. A fluidized bed gasification process development (PDU) plant of 200 kW capacity was operated with Solid Recovered Fuels (SRFs) and demolition wood. In part of the tests carried out with SRF, deposits were formed in the gas lines, and the pressure drop of the filter increased as well. However, optimal combination of gasification, gas cooling and filtration conditions were found to achieve a stable pressure drop, avoid deposit formation and to achieve efficient removal of gas contaminants. The effects of the operating conditions on the achieved carbon conversion and formation of tars and other gas contaminants were studied. The produced gas was filtered at 370-500 °C using two alternative filtration systems applying ceramic bag filters or ceramic fiber filters. The removal efficiency of waste-derived trace metals and chlorine was determined at different filtration temperatures.

1. Introduction

Air-blown circulating fluidized bed (CFB) gasifiers have been in commercial use producing fuel gas for limekilns and boilers since the mid-1980s (Kurkela, 2002). In simple fuel gas applications with clean feedstocks, the raw gases can be directly fed into the boiler or kiln without any filtration or other gas cleaning. In addition to the biomass-fueled limekiln gasifiers, two industrial gasifiers coupled to boilers have been successfully operated with waste-derived feedstocks in Finland already since the early 2000s (Wilén et al., 2004). At the Kymijärvi power plant in Lahti, Finland, a 60 MW CFB gasifier (Lahti No. 1 gasifier) was connected to a 360 MW_{th} pulverized coal fired boiler to replace 15-20 % of coal by local biomass residues and clean waste fractions. The annual gasifier availability was over 95% in each year and this plant demonstrated that CFB gasifiers are technically proven and suitable for a wide range of biomass and waste feedstocks (Nieminen et al., 2004). Another waste gasification plant, a 40 MW gasifier located in Varkaus, Finland has been in commercial operation since 2001. This gasifier was originally a bubbling fluidized bed (BFB) gasifier utilizing aluminum-containing plastic wastes as the feedstock. The feedstock originated from recycling liquid packages consisted mainly polyethylene plastics with ca. 10 % aluminum and small amounts of fiber residues. At this plant, the gasifier was operated below the melting temperature of aluminum and aluminum was removed from the raw gas using a cyclone separator before leading the gas into a gas boiler (Sumitomo, 2017). A BFB reactor instead of CFB was used in order to remove aluminum particles in metallic form, which is preferable for recycled aluminum production. In a CFB gasifier, the aluminum particles are recycled together with the bed material sand in the system, which leads to a higher degree of aluminum oxidation. Later, after the liquid package recycling plant at Varkaus was completely closed, the gasifier has been used with same type of waste from different sources. These two early examples of waste gasifiers used only cyclone cleaning before the boiler as the feedstocks were quite clean. Emission control took place after the boiler with conventional flue gas cleaning systems.

Straw and other agricultural biomass sources and waste feedstocks contain alkali and heavy metals, chlorine, and other harmful impurities, which should be removed before using the gas as fuel gas or especially when considering the production of synthesis gas. In designing gas cleaning systems for fluidized bed waste

gasification, gas filtration plays a key role in removing trace metals and chlorine from the raw gas prior to gas combustion or further cleaning (Nieminen et al., 2004). The first industrial waste gasification plant equipped with gas filtration prior to combustion in the boiler has been in operation since 2012 in Lahti, Finland (Kymijärvi II). At this plant, two 80 MW gasifier lines equipped with gas coolers and filtration units are producing gas for a 160 MW combined heat and power plant (Isaksson, 2015). This paper presents results from experimental research focused on the effects of the operation conditions and feedstock type on the performance of the CFB gasifier and on the removal of gas contaminants by dry filtration carried out at 400-500 °C. The work has been carried out to support the industrial development of CFB gasification.

2. Experimental

2.1 CFB gasification test rig

The schematic diagram of the CFB gasification PDU is shown in Figure 1. In the tests described in this paper, the catalytic reformer and final gas-cleaning units were not in use and air and steam were used as gasification agents. The gasifier had two alternative fuel feeding ports. The upper feeding point (“high”) was located approximately 2 m above the air distributor, and the lower feeding point (“low”) was close to the air distributor and thus the fuel was fed into the bottom of the bed. The gasification air was electrically preheated and could be divided into primary fluidization air as well as secondary and tertiary air feeds. Steam could be added to the fluidization air. The gasifier reactor tube (id 154 mm and total height 7.9 m), the recycling cyclone and the recycling line were electrically heated in order to eliminate heat loss. A more detailed description of the CFB PDU is presented by Laatikainen-Luntama & Kurkela (2015), where the results from gasification tests with wood and straw are described. The product gas was first cleaned in the primary recycling cyclone followed by the secondary cyclone and then the gas was led into the filter unit. Some tests were carried out without a secondary cyclone in order to increase the dust loading and specifically to lead some coarser particles to the filter to help cake build-up and removal.

Two different filter systems were used in the tests. The maximum operation temperature of the bag filter unit was 500 °C and it was designed for a low filtration velocity because the maximum pressure drop of the filter bags was limited to about 20 mbar. The maximum number of bags was 16, but typically only 12 bags were used in order to reach the targeted face velocity. The bags were located in four rows and each of them was equipped with pulse cleaning. The bags were 118 mm in diameter and 1000 mm in length. The second filter unit was designed for rigid candle filters and it contained 12 one-meter-long ceramic fiber filters. The filter elements were divided into four pulse-cleaning clusters. Both filter vessels were electrically heated to avoid further heat losses. The product gas was cooled before the filter by a heat exchanger using steam and/or water as the cooling media. The filtration systems and the used filter media are explained in more detail in Nieminen & Kurkela (2004).

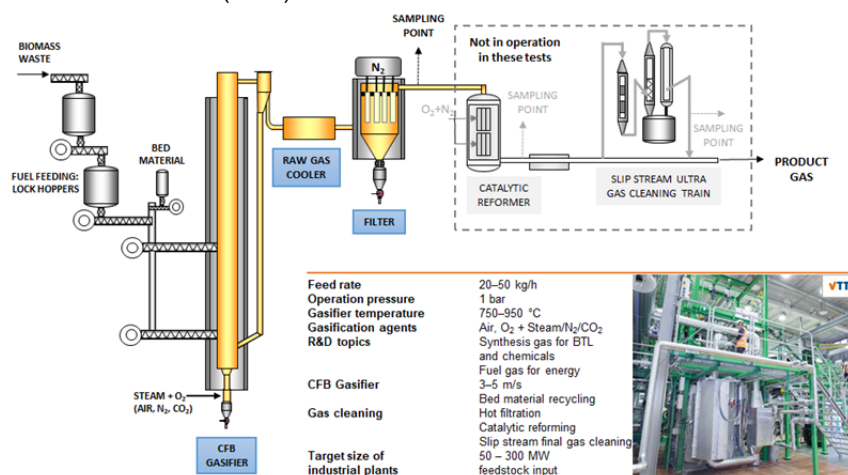


Figure 1. The CFB gasification test rig at VTT.

The product gas composition and concentrations of gas contaminants were measured from the gas line after the filter unit. The main gas components were measured using an online gas analyzer as well as a gas chromatograph. Tars, nitrogen and sulfur compounds, HCl and the concentrations of vapor phase heavy metals were measured using an extractive sampling system. The used sampling and analytical methods are explained in more detail in Kurkela et al. (1993).

2.2 Gasifier feedstocks

Table 1 below presents the averaged results for the analyses of the feedstocks used in the test campaigns. All solid recovered fuel (SRF) batches were in the form of pellets, and three different batches were used in the tests of this paper. SRF1 had a very high plastic content and thus had a low moisture content and a very high volatile matter content. SRF2 also contained some other residues but had a very low moisture content. SRF3 was like SRF2 with a higher moisture content. Demolition wood chips (DW) were crushed below 10 mm sieve before use.

Table 1: Feedstock analyses as used in the gasification campaigns.

	Moisture wt%	Volatiles wt% d.b	wt% d.b.						LHV MJ/kg d.b	
			C	H	N	Cl	S	O		Ash
SRF1	4.0	86.0	52.5	7.4	0.7	0.73	0.16	27.3	11.2	-
SRF2	5.6	77.6	52.2	7.1	0.5	0.41	0.13	31.0	8.7	-
SRF3	13.9	79.6	53.6	7.8	0.7	0.73	0.12	26.2	10.9	23.9
DW	18.7	81.5	50.0	6.1	0.3	0.02	0.03	42.2	1.4	20.1

3. Results and discussion

The results presented in this paper are from four test runs realized as 24-60-hour-long periods of continuous operation. In all test runs, the plant was operated continuously without interruptions. Measurements were carried out in 4-45-hour-long periods (set points), during which the mass flow rates of the input streams were kept as constant as possible. Elemental mass balances and performance indicators of the gasification process were calculated for the set point periods based on average measuring results. The main operating conditions and calculated performances for selected steady-state set points are summarized in Table 2.

Table 2: Main operating conditions of the gasifier at set points.

Set point	A1	A2	A3	A4	B1	B2	C	D1	D2
Feedstock	SRF1	SRF1	SRF1	SRF1	SRF1	SRF1	SRF2	DW	SRF3
Fuel moisture content, %	4.1	4.5	3.7	3.5	3.6	2.9	5.5	18.7	13.9
Fuel feed rate, g/s	8.1	11.6	4.5	9.8	7.6	9.3	9.4	7.6	6.6
Fuel feeding point	Low	Low	Low	Low	High	High	High	Low	Low
Bed additives	S+P3	S+P3	S+P3	S+P3	S+P3	S+P3	S+P3	S+P3	S+P3
Limestone feed, g/s	0.2	0.3	0.2	0.3	0.0	0.0	0.2	0.3	0.3
Sand feed, g/s	0.2	0.3	0.2	0.3	0.3	0.4	0.5	0.3	0.3
Primary air feed, g/s	14.0	14.0	11.3	14.9	12.2	13.0	15.6	14.2	14.7
Secondary air feed, g/s	0.0	5.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Steam feed, g/s	1.8	1.8	4.2	1.7	3.3	2.9	1.7	1.6	1.1
T riser - top, °C	900	906	898	851	905	863	888	887	888
Air ratio of gasification	0.24	0.24	0.34	0.22	0.23	0.20	0.25	0.38	0.34
Bed pressure drop, mbar	44	43	44	44	48	48	49	56	56
Gas flow rate, m ³ /h (wet)	81	102	73	86	78	82	85	78	72
Dry gas composition, vol%									
CO	8.8	10.8	4.7	8.2	9.7	10.4	10.6	9.1	7.4
CO ₂	13.6	13.9	14.1	14.0	14.0	13.8	14.5	17.2	14.1
H ₂	11.2	11.8	8.8	9.5	11.5	9.7	9.8	11.4	7.6
N ₂ (as difference)	57.8	53.6	66.2	57.5	55.8	54.7	55.9	58.4	62.0
CH ₄	4.56	5.34	2.94	5.24	4.87	5.54	4.77	2.80	3.50
C ₂ H _y	2.94	3.11	2.00	4.39	3.16	4.55	3.19	1.10	2.40
C ₃ -C ₅ Hy	0.05	0.06	0.03	0.17	0.04	0.14	0.06	0.00	0.05
NH ₃	0.35	0.42	0.24	0.25	0.36	nm	0.32	nm	nm
H ₂ O in wet gas, vol%	16.5	15.2	32.7	15.0	26.5	24.2	17.1	20.6	20.2
Tars + benzene, g/m ³ n	31.0	29.8	18.3	40.2	35.9	43.2	33.1	9.2	26.7
Tar + benzene yield, g/kg daf fuel	85.4	73.2	66.6	96.8	86.4	92.2	80.2	26.1	83.8
HCN content, ppm-v	45	56	-	46	161	-	92	-	-
2 nd cyclone in use	yes	yes	yes	yes	yes	yes	yes	no	no
Filter type	bag	bag	bag	bag	bag	bag	bag	candle	candle
Filter temperature, °C	412	412	412	413	445	445	394	500	500
Dust content in filter inlet, g/m ³ n	11	14	9	9	31	35	15	11	41
Filter pressure drop, mbar	7	7	7	7	6	10	10	17	19
Filter face velocity, cm/s	1.3	1.7	1.2	1.4	1.4	1.4	1.4	2.7	2.5
C-conversion, wt%									
to dry gas and tars	95.2	94.2	94.7	96.0	92.8	93.9	96.8	96.9	95.6
Determined C-losses, wt%	3.1	4.3	1.9	3.4	3.1	3.6	3.1	2.4	2.9
Mass balance closures (out/in)									
C-balance	0.98	0.99	0.97	0.99	0.96	0.98	1.00	0.99	0.99
O-balance	1.02	1.02	1.02	0.98	1.04	1.03	1.02	0.99	1.02
Ash balance	0.97	0.96	0.88	0.97	1.02	0.91	0.91	0.95	1.04

Mass balances and conversion efficiencies were calculated from the measured data in a similar way as described in Kurkela et al. (1993). In these tests, all input gas flow rates were measured, the solid input and output stream were weighed, and the composition of the feedstock and all output streams were carefully measured. The elemental nitrogen balance was used to calculate the dry gas flow rate, which could not be accurately measured. The hydrogen balance was used to calculate the water vapor content of gas. The measurement accuracy, and the obtained achievability of steady state conditions for the set points, can be judged from the carbon, oxygen and ash balances. Often the ash balance was worse due to the accumulation and release of ash from the recycling bed material especially at the set points, which were shorter than 24 hours. At some set points, elemental material balances were also calculated for chlorine, alkali, and heavy metals.

Carbon conversion efficiencies were calculated from the carbon balance with the formula: $100 \times (\text{carbon mass flow in dry gas and tars}) / (\text{carbon mass flow in feedstock} + \text{bed material limestone})$. In addition, carbon losses were defined directly from the analyses and weighing results of the removed ash streams. Conversion efficiencies were usually in the range of 94-97% and the carbon losses were found from the fly ash stream separated by the cyclone and filter. The operating conditions did not have a significant effect on the carbon conversions and already 850 °C was a sufficiently high operation temperature for the SRF feedstocks, which had high volatile matter contents.

The measured concentrations of different tar groups are presented for selected set points in Figure 2. The main tar components benzene and naphthalene are shown separately, and the “light tars” include components heavier than benzene, up to naphthalene. Toluene is the main component of this group. “Heavier tars” show the sum of concentrations of polyaromatic tar compounds heavier than naphthalene. The tar contents measured for DW gasification set point D1 are similar as reported previously for the gasification of clean wood residues by Laatikainen-Luntama & Kurkela (2015), while at the set points carried out with SRF feedstocks the tar contents were two-to-four times higher. The lowest tar contents for the SRF set points were measured at set points A3 and D2, where the equivalence ratio (ratio of air feeding in divided by the required air feed of stoichiometric combustion) was highest due to high steam feed (A3) or higher feedstock moisture content (D2). The addition of a calcium-containing bed material did not have as significant effect on the tar decomposition as reported by Laatikainen-Luntama & Kurkela (2015) for wood and straw gasification. This is evidently due to reactions of calcium with sulfur or chlorine, which may reduce the catalytic effects of CaO. The fate of the feedstock nitrogen (Figure 3) in the CFB gasification of SRF fuels seemed to follow the same pattern as has been reported for other fluidized-bed gasification tests where high-volatile biomass feedstocks are used (Leppälähti, 1998): 50-80 % of the fuel nitrogen was converted into ammonia and smaller amounts to HCN and organic tar compounds. It can be assumed that the rest of the fuel nitrogen from the material balance is converted to N₂.

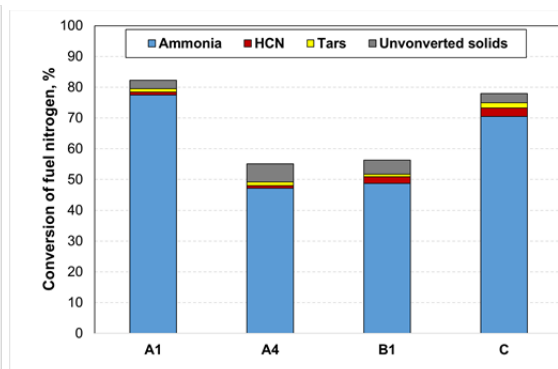
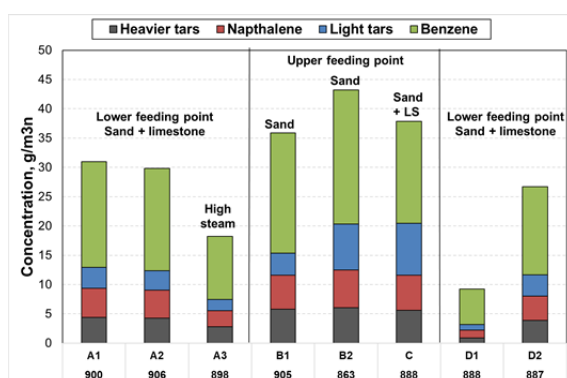


Figure 2: Concentration of tars at selected set points.

Figure 3: Conversion of fuel nitrogen to different output products at selected set points

One of the basic advantages of the gasification-based Waste-to-Energy concept is the possibility to remove contaminants, which would cause corrosion or fouling in the steam boiler or create emissions, which require complicated flue gas cleaning systems. Typical distributions of corrosive components, alkali metals, chlorine and aluminum are presented in Figure 4. Most of these elements are removed practically completely into the ash streams, while part of the chlorine passes through the filter unit as HCl. Chlorine removal of 60-80 % could be achieved just through the reactions of chlorine with the inherent sodium, potassium, and calcium present in the feedstock and in the bed additive. The chlorine removal could be improved up to 95 % by

injecting a calcium hydroxide sorbent into the raw gas prior to filtration. At the set points B1 and C illustrated in Figure 5, the Ca/Cl molar ratio was raised by feeding sorbent to 4.9-5.6, while the molar ratio was in the range 3-4, when no additional sorbent was used.

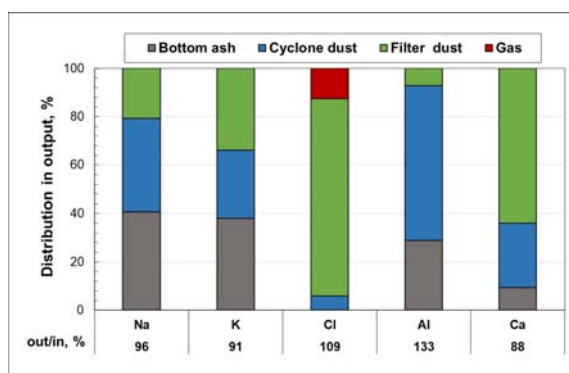


Figure 4. Distribution of Na, K, Cl, Al and Ca into different output streams (set point B1)

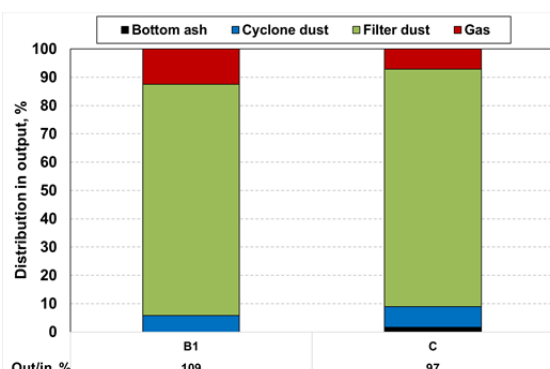


Figure 5. Mass balances for chlorine at set points B1 and C, where $\text{Ca}(\text{OH})_2$ sorbent was used.

Detailed studies on the fate of heavy metals in CFB gasification and gas filtration were carried out at set points D1 and D2. This included sampling of heavy metal concentrations after the filter unit and careful analysis of the metal contents of filter and bottom ash samples. The feedstocks were also analyzed for heavy metal contents (average results shown in Table 3). However, this was more challenging than analyzing the output streams. There was a large scatter in the heavy metal contents of individual feedstock samples. It is very difficult to take a small representative analytical sample from a large amount of rather inhomogeneous waste feedstock. The sampling of vapor phase heavy metals from hot gasification gas is also rather challenging due to very low concentrations and possibilities for sample contamination. Thus, it is necessary to compare the obtained results with those analyzed for the reference sample, which is taken from the same impinger sampling train but without passing a gas flow through the collection system. The results of Figure 6 show that only the concentrations of cadmium, lead and zinc are higher than were determined for the reference sample. Figure 7 shows that most of the heavy metals were captured in the filter ash, while the removal efficiency was somewhat lower only for cadmium. Mercury was not analyzed in this test run, as the previous tests showed that mercury is not captured by filtration at 370-500 °C, which was also verified by sampling of the gas phase mercury at set point C.

Table 3: The content of heavy metals in feedstocks of set points E1 and E2.

mg/kg	As	Cd	Co	Cr	Cu	Mn	Ni	Pb	Sb	Sn	V	Zn
SRF3	<10	<5	<10	47	197	96	14	938	26	20	<10	219
DW	<10	<5	<10	7.5	5.7	73	<10	552	<5	<2	<10	319

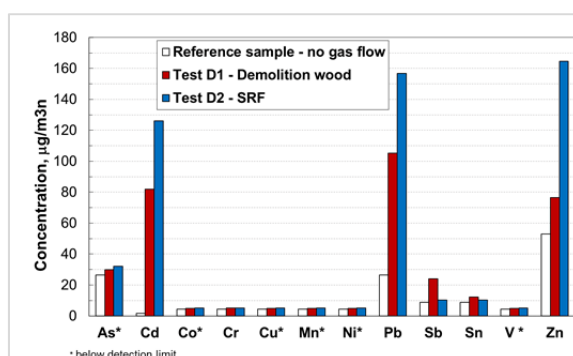


Figure 6: Concentration of trace metals in the gas after filtration at set points D1 and D2.

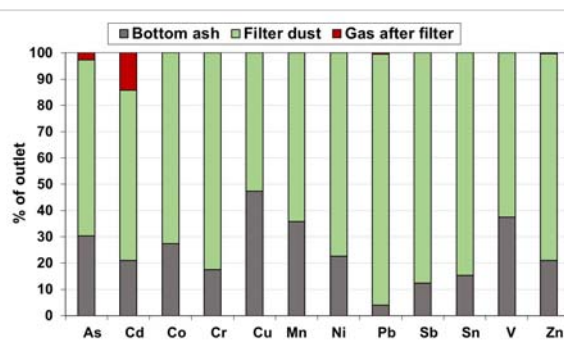


Figure 7: Distribution of trace metals in different output streams at set point D2 with SRF.

One of the key challenges of this gasification process, when operated with SRF, is the formation of sticky filter cake, which can make it impossible to clean the filters by pulse cleaning. This may result in a constantly increasing pressure drop, as was the case at set points A1, A2 and A4, where the tar content was high,

secondary cyclone was in operation and the filter temperature was low (Figure 8a). In test run A, the filter pressure drop remained at a constant level only at set point A3 where the tar content was lower due to high steam feed. Test run D illustrated in Figure 8b is an example of a more stable filter pressure drop behavior, and the change from demolition wood (set point D1) to SRF (set point D2) did not result in a significant increase in the pressure drop. In this test run, the filter was operated at ca. 500 °C and the moisture content of the SRF was higher than it was in test A. The pressure drop level in test D was higher than in other tests runs because of the higher face velocity as the candle filter unit had a smaller total filtration area than the bag filter unit.

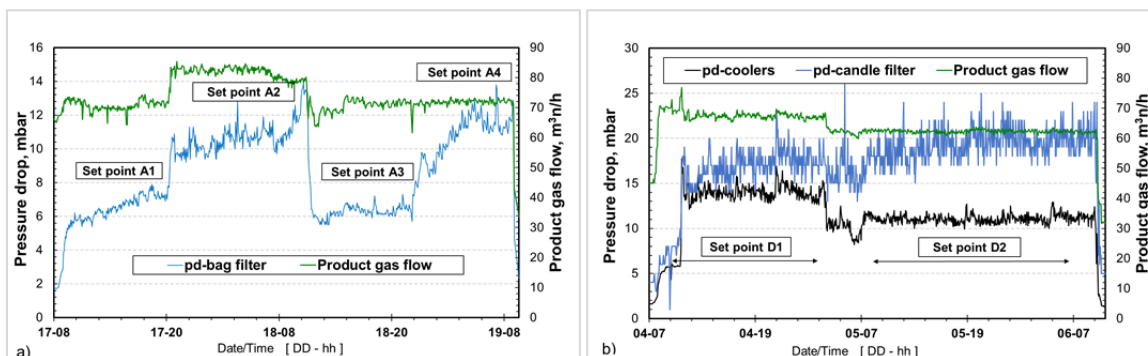


Figure 8: Pressure drops across the filter at a) set points A1, A2 and A3 and b) set points D1, D2.

4. Conclusions and further R&D

A circulating fluidized bed gasifier is a robust and feedstock-flexible reactor, which can be successfully operated with a wide range of biomass and waste feedstocks. The tar contents, however, are two-to-four times higher in the gasification of plastic-containing waste than wood gasification, and the use of calcium-containing bed material does not have a clear effect on the tar decomposition. However, the produced gas could be effectively filtered by ceramic bag or fiber filters operated at 370-500 °C. The major part of corrosive or toxic trace metals and chlorine were effectively removed by filtration. These experiments provided data and experience for designing and operating industrial scale gasification plants. Further R&D has been carried out with other types of waste materials and methods for improved management of filter ash has been developed. Presently, the same CFB test facility is also used for studying waste conversion to synthesis gas via steam/oxygen gasification followed by catalytic reforming.

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