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RDF-Gasification part 1: Characterizing the use RDF as fuel and solving the tar problem by an in- depth laboratory study

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RDF-GASIFICATION PART 1: CHARACTERIZING THE USE OF RDF AS FUEL AND SOLVING THE TAR PROBLEM BY AN IN-DEPTH LABORATORY STUDY.

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SUMMARY: Source separation of waste and higher recycling percentages in waste streams result in better sorted waste, e.g. in RDF with a high plastic content. An optimized, dedicated installation can be designed for this high calorific waste. Preliminary laboratory tests showed that gasification of RDF has some advantages over combustion in a fluidized bed system. The drawback of gasification, however, is the production of tars. The tar chemistry was studied during laboratory-scale gasification tests at ECN with emphasis on RDF specific tars and their effect on the OLGA tar-cleaning system designed for biomass. The first tests encountered polymerization in the OLGA collector. The influence of foam polystyrene in the fuel was studied after observing extremely high styrene concentrations. However, polystyrene by itself did not lead to fouling. A new hypothesis is currently experimentally tested. It is thought that the polymerization can be attributed to the presence of polystyrene in the RDF fuel, chlorine as a catalyst for the polymerization reactions and at least one other tar component, likely released from another RDF component. Future efforts will be aimed at understanding the influence of the main individual RDF components and building up a knowledge base to use as a predictive tool to avoid operational problems.

1. INTRODUCTION

Source separation of waste and higher recycling percentages in waste streams result in better sorted waste, e.g. in RDF with a high plastic content. Instead of burning this fraction in a general Municipal Solid Waste (MSW) Incinerator, where it often will cause problems when not properly mixed, a more optimized dedicated installation can be designed for this high calorific waste. Preliminary laboratory tests showed that gasification of a relatively high calorific value fuel, such as RDF, has some advantages over combustion in a bubbling fluidized bed system. Especially the feeding of the fuel in the bed was very problematic under combustion conditions while this problem was clearly less problematic when gasifying the fuel. In addition, the efficiency of the gasification process is higher and the use of the product gas allows a more versatile end-use than steam generation only, for example by upgrading towards a synthetic natural gas or a transportation fuel. The drawback of gasification, however, is the production of tars

Tar related problems are the most important obstruction for the introduction of gasification and are responsible for major operational problems due to tar condensation (fouling) and poisonous condensate leading to unacceptable waste water costs. Over the past decade, intensive

research lead to the development of the OLGA cleaning technologie.

OLGA is an advanced scrubbing technology for tar removal that was developed and patented by ECN. Dahlman Industrial Group BV, Maassluis (the Netherlands), brings the technology to market. The technology does not concentrate on the tar quantity, but on the unwanted behavior of tar (i.e. the properties). The performance of OLGA was demonstrated by a 1000 hours duration test with a 0.5 MW(t) pilot plant at ECN. The process showed to be stable and reliable and gas quality was on specification for a gas engine that ran smoothly. Results on tar dew point (<10 °C), phenol removal (completely) and naphthalene removal (>99%) were excellent. Currently the technique is being demonstrated in France on a 4 MW(t) scale gasifier.

The above described 1000 hours success story was performed using clean wood as fuel. Whether or not similar behavior can be expected when the fuel was not clean wood but a difficult waste fraction such as RDF is not known. Because the detailed characterization of the tar content depends on very stable and well known operating conditions it was decided to start work on RDF in a lab-scale bubbling fluidized bed with a lab-scale version of OLGA, as was done at the beginning of the biomass work. A test program was performed of which the results will be presented. Furthermore, the RDF fuel used in test series 1 was also tested in a pilot-scale installation in Tzum (Netherlands) as a prelude for a commercial scale gasifier-OLGA installation (now in the procedure of permissions) that is planned to use the same RDF. (See presentation "Gasification of RDF part 2: Pilot-scale results").

2. TAR CHEMISTRY AND MEASUREMENT METHODS

2.1 Tar chemistry versus tar quantity

Tar formation during gasification is not avoidable when using fluidized bed technology at temperatures typically below 900 °C. Tar is formed due to the depolymerization of the building blocks of biomass (e.g. wood and paper) and the plastic components in RDF. The term 'tar' includes thousands of single substances. A general definition of 'tar' often used is: all organic contaminants having a molecular weight higher than benzene.

It is widely recognised that tars are the most problematic species in product gas derived from biomass gasification. Tars easily lead to fouling and largely depends on the tar dewpoint and composition of the tars and not so much on the total tar concentration. This is shown in Figure 1. For example just some tar components that have 4 or more aromatic rings raise the dewpoint of the productgas from 120 °C to more than 200 °C, bringing it in the temperature range of gas cleaning components.

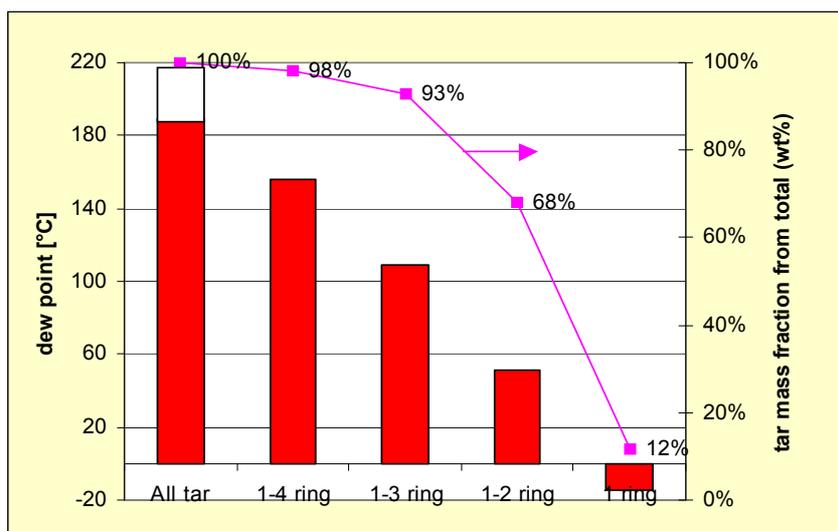


Figure 1: Dewpoint versus mass fraction of tars with different amounts of aromatic rings. The red bars show the dew point temperature depending on the composition. The pink line shows the total amount of tars as indicated on the Y-axis on the right hand side.

2.2 Tar sampling methods

There is a tar guideline sampling and analysis method that is the CEN standard and is often used at ECN both in the lab and during commercial scale measurement campaigns. However, it requires long stable operation conditions and in addition solid-phase adsorption (SPA) is often used for its quick and easy application. Because the lab-scale experiments are performed under frequently varying conditions, SPA was the method of choice. The method comprises of ≈ 100 ml of hot producer gas being passed through a disposable cartridge containing 500 mg of amino-phase (modified silica). After preparation, samples are analysed on a chemical lab for individual compounds by GC-FID/MS. Its main advantage is the easy and fast sampling (2 minutes). Disadvantage is that only GC detectable components ranging in molecular weight from 78 (benzene) to 300 (coronene) are determined. The method is less suitable for the very volatile tar components like benzene and toluene. Experience is that there is an overall good fit between results of the SPA method compared to the Guideline method for most of the tars and where the concentrations of SPA are (much) lower in the range of the volatile components, the values are at least reproducible and are used as a rough first qualitative indication (See e.g. “Gasification of RDF part 2: Pilot-scale results”).

3. EXPERIMENTAL LABORATORY STUDY

3.1 Aims

The first aim of the experimental program was to test RDF as a fuel under gasification conditions in a fluidized bed reactor. The feeding of RDF, the bulk gas conditions and the formation of tars were aimed to be studied, but also the behaviour of the fly ash and the potential to agglomerate the bed material (quartz sand). Due to the enormous variety of RDF it was also aimed to try at least a few different streams of RDF. In case gasification would be successful, the aim of the experiments would further focus on the gas cleaning, necessary to bring the gas up to

specification for its end use. In particular experiments were designed to test the suitability of the OLGA tar removal system for a fuel such as RDF.

3.2 Apparatus

The experimental set up is shown in Figure 2 and consists of the bubbling fluidised bed (BFB) reactor "WOB" on the left with subsequently a cyclone and a hot gas filter for solids removal, the columns (collector and absorber/stripper) of the Olga tar cleaning system and the wet scrubber on the right side of the drawing after which the cleaned gas is sent to the flare.

In general, OLGA is operated downstream of a low-efficient solids removal step (e.g. a cyclone) and a wet electrostatic precipitator between collector and absorber. However, in the lab-scale set up the a high efficient solids removal step (i.e. a hot gas filter) is applied upstream OLGA. In the OLGA the product gas is cooled with oil in the collector, where the liquid tars are collected. Further downstream, gaseous tars are absorbed in the scrubbing liquid of the second column

The liquid tars in the collector are separated from the scrubbing liquid and returned to the gasifier. A small amount of the scrubbing liquid is bled and recycled to the gasifier. For the absorption step, scrubbing columns were selected that are interacting with each other in a classical absorption-regeneration mode. The scrubbing liquid from the Absorber with the dissolved tars is regenerated at high temperature in the Stripper. In case of air-blown gasification, air is used to strip the tar. Subsequently, the air with the stripped tars is used as gasifying medium. The loss of scrubbing liquid in the Stripper by volatilisation is minimised by the use of a condenser.

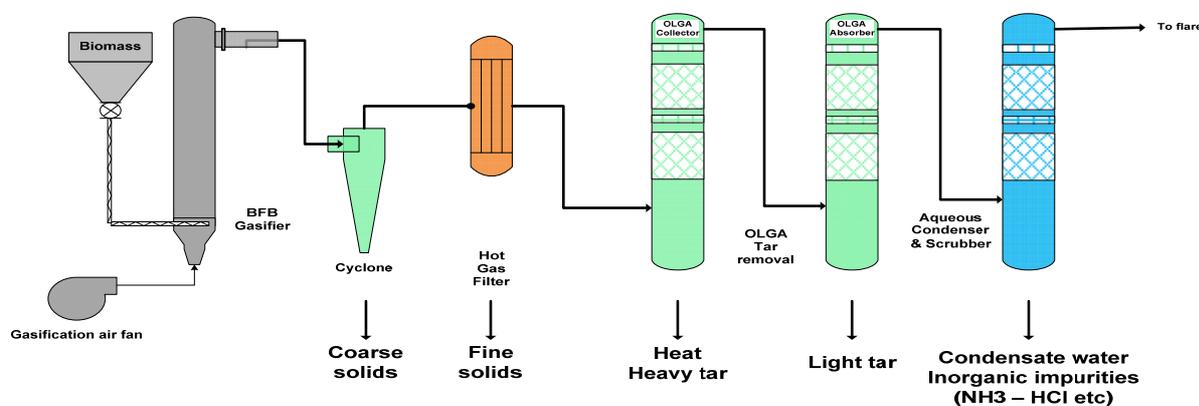


Figure 2. Process flow diagram.

3.3 Experimental procedure

Three different test series gasifying RDF were run in March, May and June 2008. The first series was aimed at establishing the gasifier optimum conditions and deal (when necessary) with feeding problems. To make best use of the experiments the OLGA collector was in operation and tar characterisation using SPA sampling was performed in case stable enough operating conditions could be established. Series 2 and 3 were aimed at testing different RDF streams as well as operating the full gas cleaning train and characterizing the gas and tar composition after each component in the experimental set up. Due to unexpected polymerisation problems the aims

of series 2 and 3 were slightly modified, as will be explained in Section 4 describing the results. Test series 1 results led to a more fundamental procedure change as well, also for future work. Instead of testing different streams of RDF all over again, the efforts are aimed at truly understanding the influence of the main individual RDF fuel components and building up a knowledge base. Series 2 with the expanded (foam) polystyrene (EPS) fuel is an example of this.

For the lab-scale installation the RDF fuel, delivered as chips, is first dried overnight at 85 °C, then milled under liquid nitrogen and sieved to a maximum size of 2 mm. The size of the fuel particles is mainly restricted due to the small screw feeder that transports the fuel from the bunker into the bed of the gasifier. The initial moisture content is re-introduced in the form of steam in the gasifier. For all tests the steam was set equivalent to 25% of moisture in the fuel and the additional air for the gasification process was calculated to result in an equivalence ratio (ER) of ~0.35. The amount of fuel fed in the reactor was roughly 500 g/h and always checked afterwards by weighting the remaining fuel. Two principle test temperatures were used, i.e. 850 °C as the temperature conversion temperature for waste incineration and 750 °C as the lower temperature alternative, also used in the pilot scale tests in Tzum. The higher heating value of the RDF was roughly 25 J/g.

In Table 1 the different fuels of the three test series are given, the temperatures of gasification and some remarks on the test itself.

Table 1. Fuels, test conditions en remarks on the performed experiments.

Test series	Dates	Fuel	WOB temp.	Remarks
1	27th of March –April 16th 2008	<ul style="list-style-type: none"> RDF1 and RDF2 from demolition and construction waste (high EPS/0.23 wt% Cl and high EPS/ 0.38 wt% Cl) 	750°C and 850°C	Focussed on gasification proces and feeding (bunker problems). Polymerisation problem in OLGA collector
2	21-23rd May 2008	<ul style="list-style-type: none"> RDF3 from recycler (low EPS/1.8 wt% Cl) EPS10% mix with wood (no Cl) 	750 °C and 850°C	RDF3 experiment steam forgotten in first part. Low EPS/high Cl EPS tested as single RDF component, no Cl
3	11-12th June 2008	<ul style="list-style-type: none"> RDF4 from household packaging waste minus EPS/ 1.1 wt% Cl 	750°C and 850°C	RDF4 without EPS, complete smooth operation

4. RESULTS

4.1 First test series

During the first test days several feeding problems and plugging problems occurred. Reconstruction of the outlet piping of the WOB was necessary due to plugging with ash in horizontal parts of the piping and in sharp bends. Also the small particles < 0.7 mm were sieved out of the fuel. After these actions most problems were gone although stable feeding of this inhomogenous material stays difficult at lab scale and many adjustments of the fuel feeding system and bunker were done during this test series.

During the first test series it was established that no problems considering agglomeration of the sand bed took place. Limited amount of ash (8%) accumulated in the bed section, roughly 50% was collected in the cyclone and 26% in the hot gas filter. The remaining 16% was present after the filter but reduced to $0 \pm 10 \text{ mg/m}^3$ after the OLGA collector. In Figure 3, the temperatures measured at various locations in the gasifier are shown with time. The irregular pattern shows the difficulties with the feeding system. Similar graphs of test series 2 and 3 show a more smooth behaviour. The same feeding problems are reflected in the bulk gas and minor gas compositions shown in Figures 4 and 5. The gases were measured by on-line analysers and micro-GC.

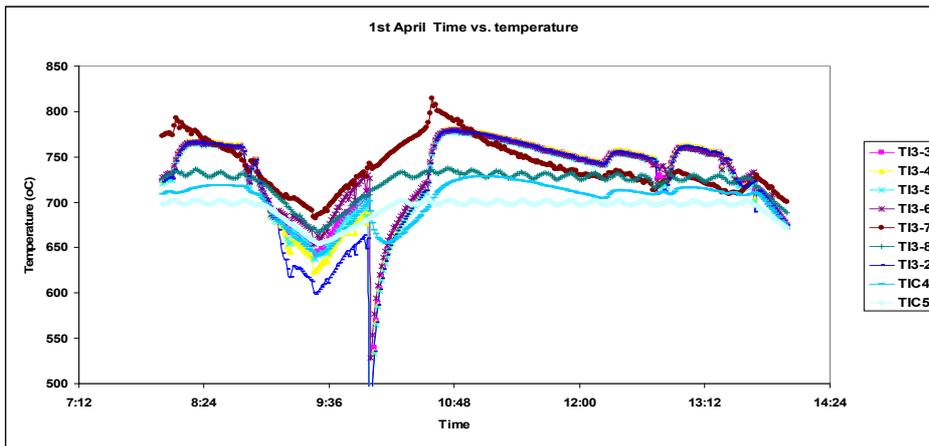


Figure 3. WOB temperature measurements with time.

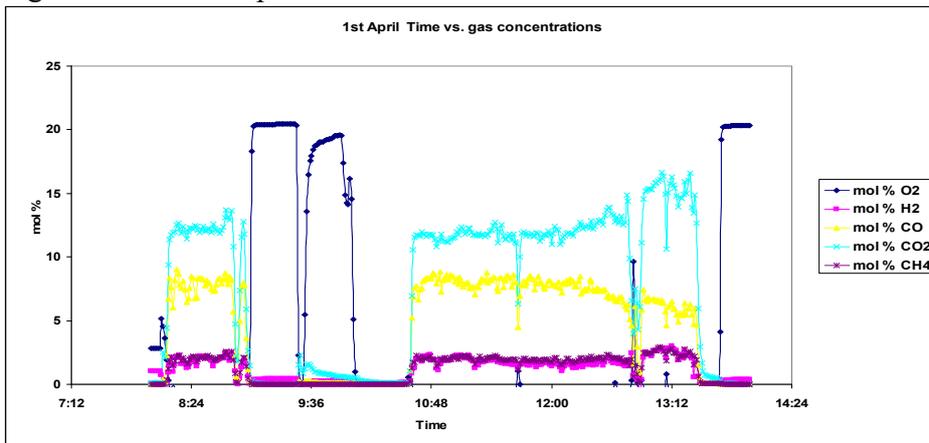


Figure 4. The main gas concentrations of the producer gas after the WOB with time. N_2 is excluded to benefit the scale of the other gas concentrations. The few high oxygen intervals show the problems with the feeding system

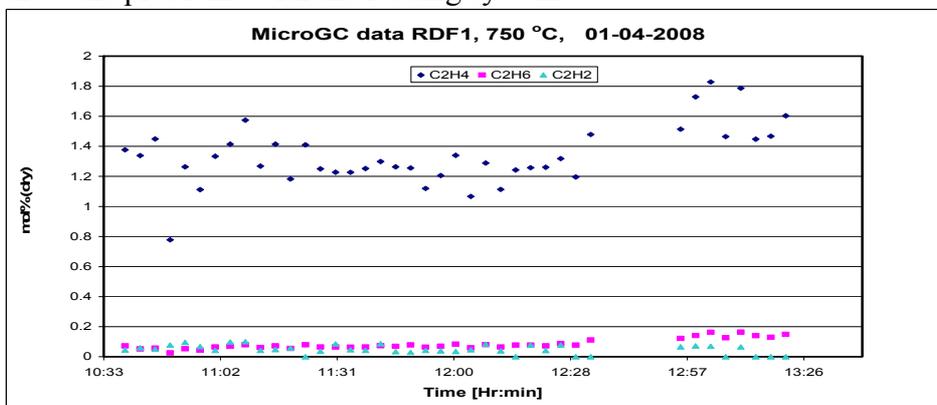


Figure 5. Minor gas concentrations, i.e. C_2H_4 , C_2H_6 , C_2H_2 in the producer gas with time.

SPA tar sampling was performed only if gasification lasted at least 1 hour.. The concentration is lower after the collector than before, which is unusual for a volatile tar. It is very typical fuel related peak not seen in this concentration with any of the biomass streams tested in the past. Naphtalene is the next highest peak but shows a reverse behaviour with the higher concentration after the collector

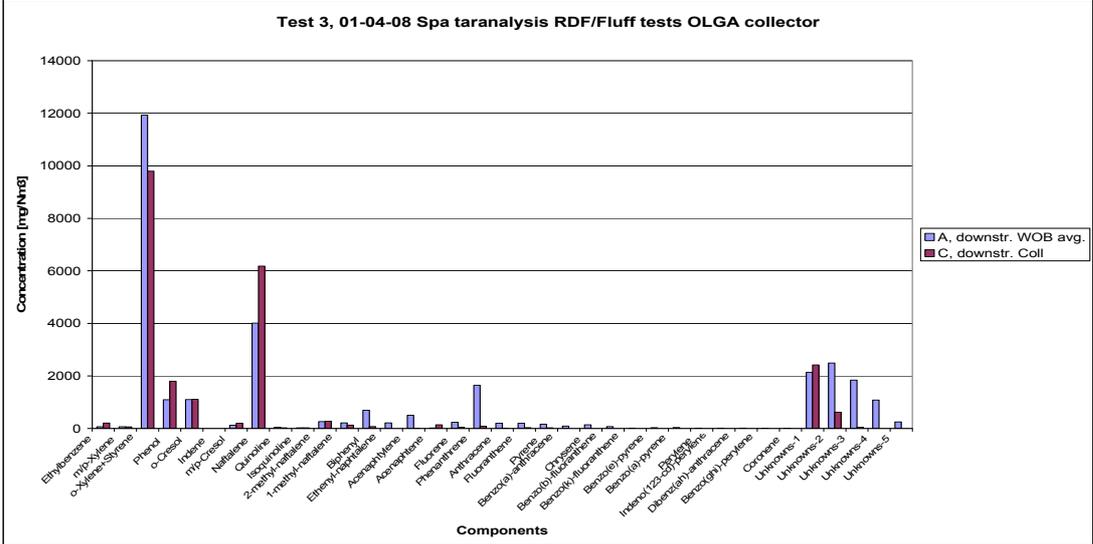


Figure 6. Tar concentrations in the productgas before and after the OLGA collector column

During all the tests in series 1, a significant and unusual temperature increase was registered in the upper sections of the OLGA collector column (except for the restricted exit temperature). Also the pressure drop over the collector column increased and testing had to be stopped. The increase occurred faster with higher chlorine contents (RDF2) and at lower gasification temperature (750 °C). The temperature increase is shown in Figure 7 by the light blue line as an example of one section. The temperature raise is thought to be a result of an exothermal reaction.

The OLGA collector was dismantled and the different sections were inspected on fouling. The lower sections were clean, the upper 3-4 sections showed some fouling with black and sticky material, thought to be polymerized tars produced during the exothermal reaction.

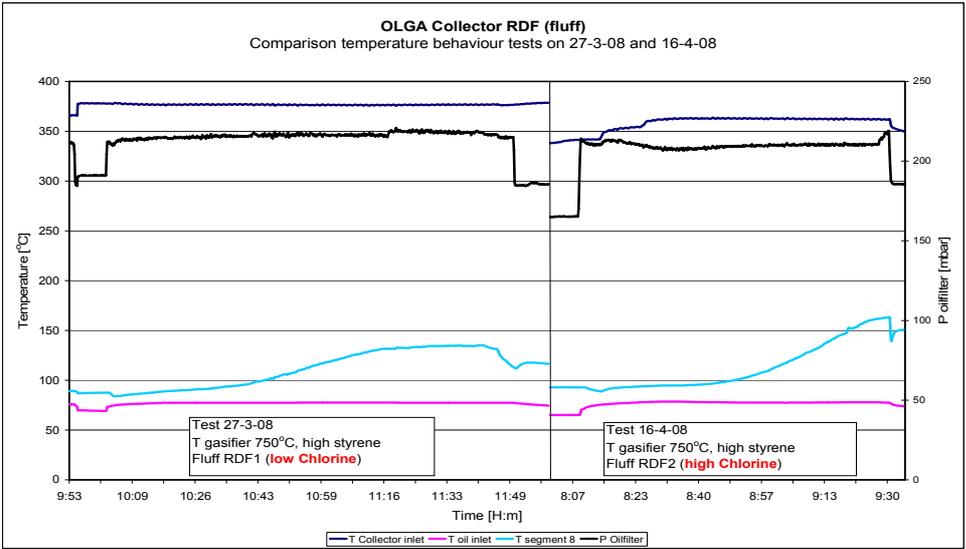


Figure 7. Collector temperature.profiles and oil pressure, RDF1 test1 (27-3) and RDF2 test 7 (16-4). The dark blue line and the pink line represent the fixed in- and outgoing temperatures, respectively.

On the black fouling deposit, sampled from the OLGA collector some first analyses were performed. Some preliminary FTIR analyses showed the main component to be a rubber-like substance. More detailed analyses will be performed on the deposit sample soon.

4.2 Second test series

The extremely high styrene peak in the first series, related to the RDF fuel, which was reduced after the collector, the exothermal (polymerization) reaction likely to have resulted in the fouling as well as the aggregating effect of chlorine led to the approach of the second series to test the EPS component separately and without chlorine and to choose an RDF high in chlorine and low EPS. RDF3 was a milled sample without any visible EPS, however, the styrene peak during the test run suggested it was not very low in EPS. A better choice, RDF4 was chosen for test series 3. Here the results of the EPS test are described. Figure 8 shows the bulk gas composition as an example of the smooth operation. In Figure 9 the light blue lines show again the exothermal reaction, although no fouling was established. It shows EPS is an important factor in the problem but its presence alone is not enough to cause the fouling.

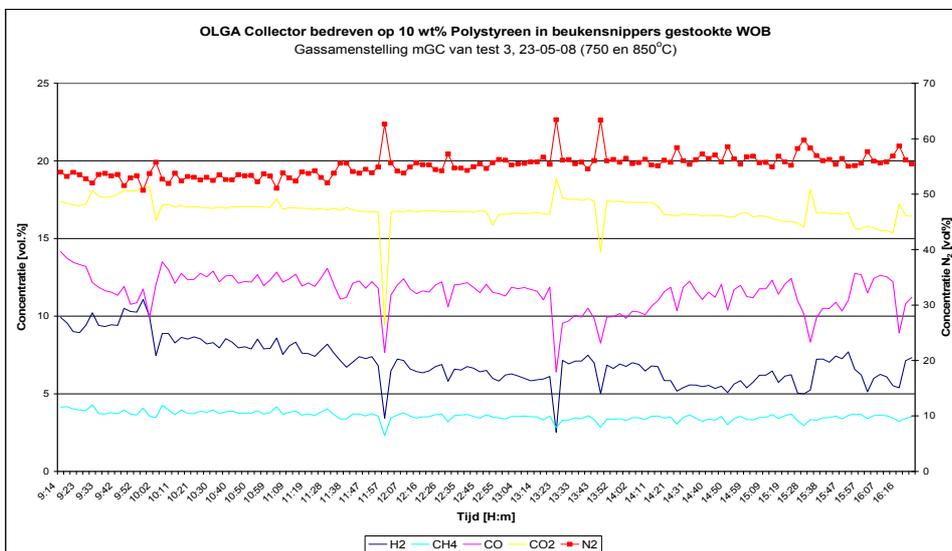


Figure 8. The main gas concentrations of the producer gas after the WOB with time. N_2 is on a separate scale on the right axis.

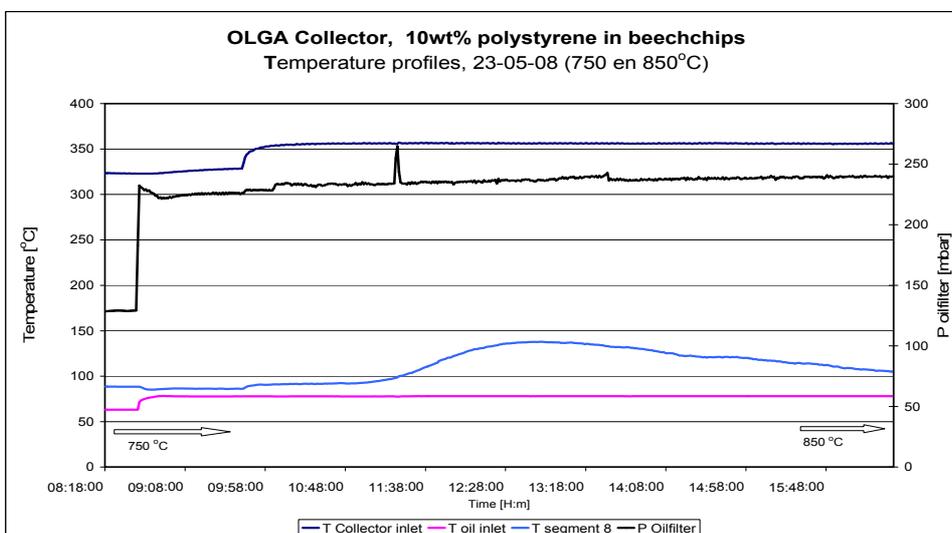


Figure 9. Temperature profiles of the collector column test 3 on May 23 rd.

4.3 Third test series

On the 11th and 12th of June experiments were performed using RDF4 from household packaging waste, separately collected by Hummel Recycling, from which all visible EPS was removed. However, this type of packaging waste contains other forms of solid (non-foam) polystyrene which remained present. The test did run smoothly as can be seen from Figure 10 showing the bulk gas composition. The tar composition, sampled before and after the collector of OLGA (Figure 11) shows that no styrene remains in the collector. Otherwise, most of the tars are much higher before the collector (in blue) compared to the concentrations after the collector (in purple) showing a very good performance in tar reduction. No temperature rise in the upper sections of the collector could be detected (see Figure 12) and no fouling took place.

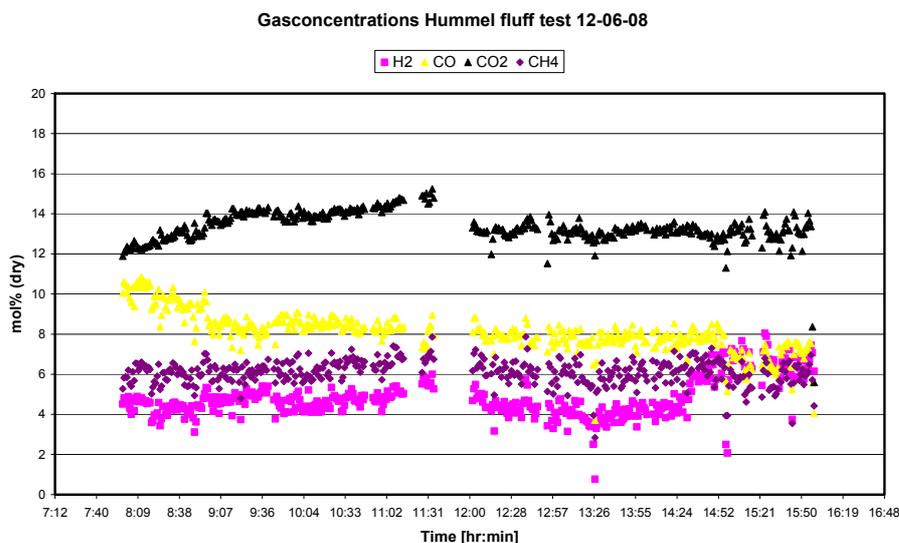


Figure 10. The main gas concentrations of the producer gas after the WOB with time. N₂ is excluded.

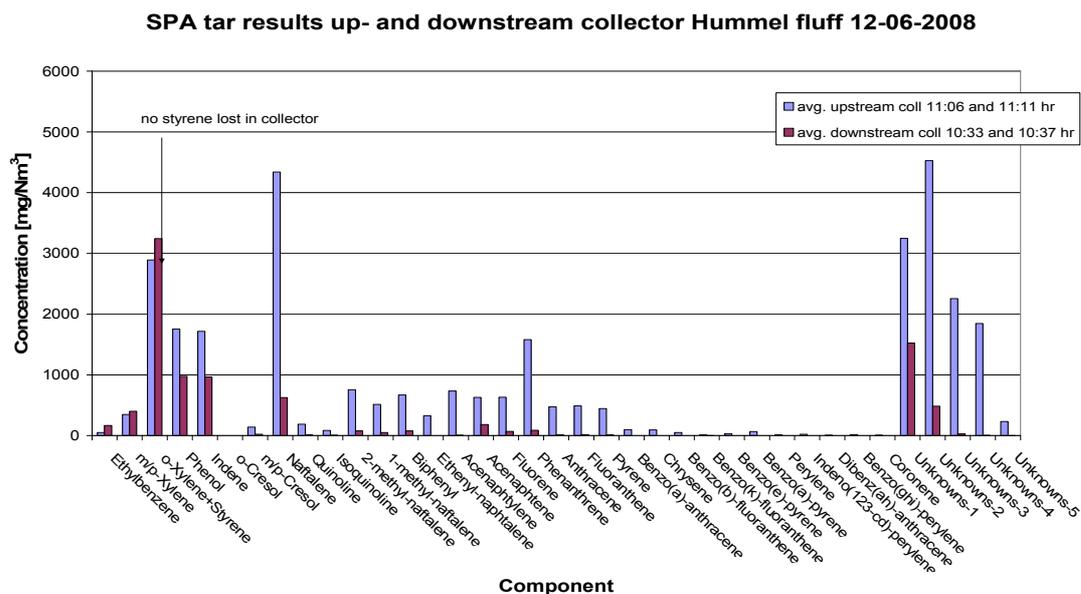


Figure 11. Tar concentrations in the product gas, before and after the OLGA collector.

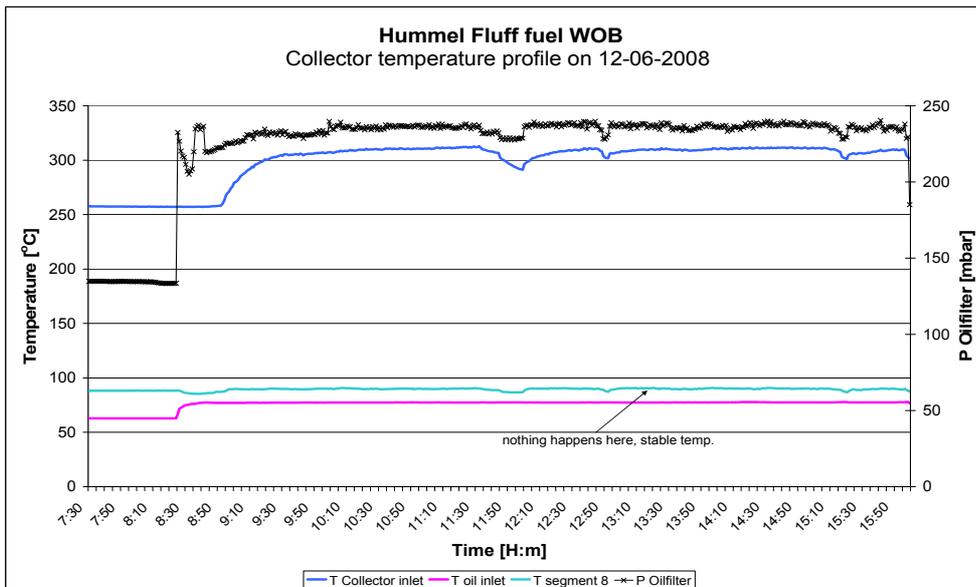


Figure 12. Temperature profiles of the collector column on June 12th. Note that the light blue line does not indicate at an exothermal process.

5. DISCUSSION AND CONCLUSIONS

Despite some feeding and plugging problems during the first test series, a generally good performance of the fluidised bed gasifier on RDF was shown. The bulk and minor product gas composition with time show a stable trend. No signs of agglomeration of the sand bed were observed. Overall, the application of a fluidised bed as dedicated installation for RDF gasification is promising.

Focussing further on the tar removal system, optimization of the OLGA is needed because the tar concentrations are different for RDF than for the biomass fuel for which it was designed. Sample analyses during the experiments on the product gas showed that the most prominent peak in the tar concentrations is that of xylene/styrene. Cleaning the gas from the gasifier with OLGA, using RDF as a fuel, resulted in a fouling problem in the OLGA collector column during the first test series. The temperature rise was probably caused by an exothermic reaction. These problems occur faster gasifying at 750 °C than at 850 °C. Other RDF from household packaging waste without expanded (foam) polystyrene (EPS) did not give any problems. The polystyrene itself used as a fuel in 10% mixture with beech did not result in fouling problems either.

The formation of a rubber-like substance within the gas cleaning section seems related to the plastics in RDF. A typical composition of RDF contains among others polyurethane foam (PUR), polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), acrylonitrile butadiene styrene (ABS) and polystyrene (PS). By gasifying the RDF material, these components will decompose into the individual building blocks ethylene, propylene, butadiene and/or styrene, which via chain reaction polymerization can form an heterogeneous mixture of different difficult to identify thermoplastic polymers, although polymerization of butadiene and styrene is more likely to occur than of propylene or even ethylene.

A new hypothesis will be experimentally tested in future work. It is thought that the polymerization can be attributed to the presence of polystyrene in the RDF fuel, chlorine as a catalyst for the polymerization reactions and at least one other tar component, likely released from another RDF component. Future efforts will further be aimed at understanding the influence of the main individual RDF components and building up a knowledge base to use as a predictive tool to avoid operational problems.

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