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## Tar Formation and Destruction in a Simulated Downdraft, Fixed-Bed Gasifier: Reactor Design and Initial Results

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A suite of improved technologies is being developed to minimize the environmental impact of biomass/waste fired gasification processes. Downdraft, fixed-bed reactors are particularly favored because of their ability to destroy the majority of tars produced from the fuel volatiles. However, there is some concern about the impact of the low residual tar concentration on the long-term operational reliability. A two-stage laboratory scale fixed-bed reactor has been constructed for studying the release and destruction of tars in downdraft gasifiers. The reactor has been commissioned and its performance demonstrated using several biomass feedstocks. Experiments using the first stage only have shown that as the temperature is raised from 250 to 450 °C, the gas and tar yields increase at the expense of the char residue. Four different biomass/waste materials (eucalyptus wood, sludge, plastic waste, and silver birch wood) showed qualitatively similar behavior. Volatile yields appear to stabilize around 450 °C. With silver birch wood, the tar yield reached 47% of the initial fuel. Preliminary tests using a char bed in the second stage have been completed. The presence of the throat and the second-stage char bed results in a substantial reduction in the quantity of tar leaving the reactor. With a hot empty second stage (at 800 °C), the tar content was reduced to 5.3% (by wt of initial fuel charge) in the exit gas from the reactor. Packing the second stage with char (at 800 °C) further decreased the tar content to less than 0.1%. Gas analyses have been performed, showing that some of the initial tar is broken down mainly to CO and CH<sub>4</sub> in the second stage of the reactor. Further work is in progress to study the impact of the operating conditions in the second stage on the residual tar concentrations and gas analysis.

### Introduction

Carbon from gaseous, liquid, and solid fossil fuels reacts with oxygen from the air during combustion and thereby releases fossil derived CO<sub>2</sub> into the atmosphere. This anthropogenic release is contributing to a rise in atmospheric CO<sub>2</sub> concentrations. There are serious concerns about the impact of this increasing concentration in the earth's atmosphere on the world climate, through an enhancement of the greenhouse effect. On a worldwide basis, the use of fossil fuels is increasing, with large increases in usage in countries with developing and transitional economies, in addition to present and projected increases in already industrialized countries. One way to limit the rising concentration of CO<sub>2</sub> in the atmosphere is to use biomass and organic wastes as a replacement fuel for coal. These biofuels are mainly comprised of carbon, oxygen, and hydrogen,<sup>1</sup> but the difference between these and the fossil fuels is that biomass is part of a natural, short-term cycle and is not considered as resulting in a net increase in the amount of carbon in the atmosphere.

There are many sources of biomass and waste fuels. The term biomass generally refers to material that is deliberately grown as an energy crop. However, energy crop plantations have fundamental long-term problems. Cultivation of crops for fuel depletes the soil of nutrients, which have to be replaced. One widely applied nutrient is fertilizer grade phosphate rock, but the reserves are limited, with 400–500 years of this resource

remaining at present rates of consumption. The growing of trees, for coppicing as a fuel, in marginal lands depletes already marginal soil. Between 2500 – and 3300 ha are needed for a 10 MWe (megawatt electrical) output in contrast to cultivation for higher value end products, such as sugar cane, beets for sugar, opium for pharmaceuticals, or vegetables and cereals for food. The same area of land could feed approximately 7500 people and 850 head of cattle.<sup>2</sup> Therefore, there is a strong case for maximizing the use of existing biomass and waste materials as fuels rather than deliberately growing energy crops. This route is more economic, as well as providing significant environmental benefits. A reduction in the volume of waste for disposal is achieved by the utilization of these materials. This will reduce the waste disposal costs incurred by industry and communities, which currently amount to approximately \$60 tonne<sup>-1</sup> for non-hazardous materials.<sup>3</sup> These charges will rise as landfill sites become increasingly scarce and expensive, as a result of more stringent environmental legislation.

Gaseous and liquid fuels, chemicals, power, and heat can be produced by the thermochemical conversion of biomass and waste materials. In the biorefinery concept, it is possible to utilize biomass to generate a number of product and revenue streams that could revitalize rural economies, increase national security by reducing the dependence on oil and gas imports, and improve the global environment by reducing fossil fuel usage. There is a wide range of process options for converting

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(1) Turare, C. *Biomass Gasification Technology and Utilization*; ARTES Institute: University of Flensburg, Germany, July 1995.

(2) Kandiyoti, R. Bench scale experiment design for developing co-pyrolysis and co-gasification technologies. *Int. J. Power Energy Syst.* **2004**, 24, 205.

(3) U.K. Government Energy White Paper. *Our Energy Future Creating a Low Carbon Economy*; DTI/pub URN 03/660, February 2003.

biomass and wastes into usable fuels. These are broadly divided into pyrolysis to produce a mixture of solid char, condensable liquids (bio-oil), and gases; combustion for heat and power generation; and gasification to provide a fuel gas for subsequent combustion. Biomass gasification is not a new technology, but there is a renewed interest in its further development, mainly to produce power and heat as part of locally based combined heat and power plants.<sup>4</sup> Development is needed to ensure that the efficiency of the process is maximized and that the environmental impact is minimized. One very widely considered technology for this application is the downdraft, fixed-bed gasifier.<sup>5</sup> However, tar formation can occur, although at much lower levels than those found with updraft fixed-bed systems. Tars formed during the gasification of biomass are a complex mixture of many compounds and may contain thousands of individual compounds, with a wide range of boiling points. The composition and amount of tars depends on several factors, such as type of gasifier, moisture, and size of biomass, and operating conditions, such as pressure, temperature, and residence time. Some problems caused by tars in biomass gasification systems are condensation of heavier tar components on cooler surfaces and formation of aerosols, formation of soot by the polymerization of tars in the gas phase, and interaction with other contaminants (e.g., by adsorption on fine particles). These problems can lead to a blockage of particulate filters and clogging of fuel lines and injectors in internal combustion engines. Therefore, gas conditioning is required to remove these unwanted species, and this generally involves an integrated, multistep approach that depends on the end use of the product gas.<sup>4</sup>

In the present study, a two-stage hot-rod fixed-bed reactor has been constructed to simulate conditions in a downdraft gasifier. Experiments are being conducted to study tar destruction and how the process operating conditions affect the tar yield from different feedstocks. The main objective is the identification of conditions for the production of tar free gas from downdraft gasifiers. In this paper, the development of the equipment and initial results are described.

## Materials and Methods

The initial development of the hot-rod reactor configuration goes back to work at the U.S. Bureau of Mines.<sup>6</sup> An account of the successive stages of development of this type of reactor since then has been described elsewhere,<sup>7</sup> and the evolution of the hot-rod reactor design at Imperial College has been outlined in several publications.<sup>8–12</sup> The configuration of the single-stage atmospheric pressure hot-rod reactor used for part of this study is shown in Figure 1. The reactor body used in experiments described next was

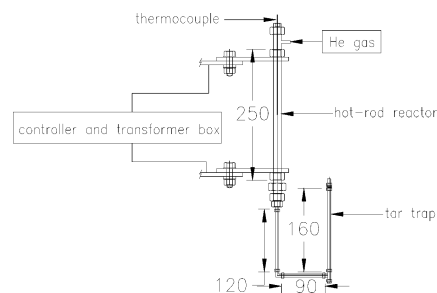


Figure 1. Single-stage hot-rod fixed-bed reactor.

250 mm long with a 12 mm internal diameter, constructed of AISI 316 stainless steel.

The top of the reactor was fitted with a T piece, connecting the gas inlet supply and allowing an internal reactor temperature measurement using a thermocouple. The tar trap at the outlet (bottom) was connected to the reactor via a reducing union. This trap was immersed in liquid nitrogen and used to capture volatiles released by the sample during the reaction. The reactor was resistance heated, and the temperature was controlled by means of the internal thermocouple. Two thermocouples attached to the external reactor wall were used for monitoring temperature excursions. These thermocouples were connected to a PC for control purposes and for data acquisition. The initial electrode design has been discussed elsewhere.<sup>13</sup> The original reactor body was attached with fixed rigid electrodes at the top and bottom of the body. When the peak operating temperature of the reactor exceeded 1000 °C, however, thermal expansion became significant, and several reactor bodies were lost through distortion, from being held between fixed electrodes. The solution adopted was from an earlier design of a fluidized-bed reactor, where similar problems had been encountered.<sup>14</sup> In the present design, the top electrode was connected to the transformer with copper bars, which served to support the reactor in position. However, the bottom electrode was movable (floating), to accommodate the thermal expansion of the reactor body and was connected to the transformer with flexible copper cables.

The operating conditions for the single-stage reactor were as follows: heating rates up to 10 °C s<sup>-1</sup>, an operating temperature up to 800 °C, a pressure up to 150 bar (or 40 bar at 1000 °C), and sample sizes of up to 1 g. The heating rate was limited to a maximum of 10 °C s<sup>-1</sup>, to ensure the absence of radial temperature variations.

**Construction of a Second Stage with He/Air Injection at the Flange.** A second reactor stage with He/air injection at the flange was added to the base of the existing reactor (pyrolysis stage, shown in Figure 1) to simulate the throat and reducing bed of a large scale reactor. In the laboratory simulation, the second stage incorporated a section where diluted air may be added, which was followed by a char bed, mainly under reducing conditions. Undiluted air cannot be added in the laboratory reactor because of potential overheating problems. The design of the heating electrodes of the second stage was the same as used for the first stage, except that both electrodes were mounted flexibly.

A two-stage reactor had previously been used in this laboratory to produce tars in the pyrolysis (top) section, for subsequent hydrocracking over a fixed-bed catalyst in the second stage.<sup>15</sup> In the new version, the design concept is similar; however, a throat/gasification section has been adopted, and the gas stream added to the throat may be altered between inert gas and diluted air injection.

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(9) Bolton, C.; Snape, C. E.; O'Brien, R. J.; Kandiyoti, R. Influence of carrier gas flow and heating rates in fixed bed hydrolysis of coal. *Fuel* **1987**, *66*, 1413.

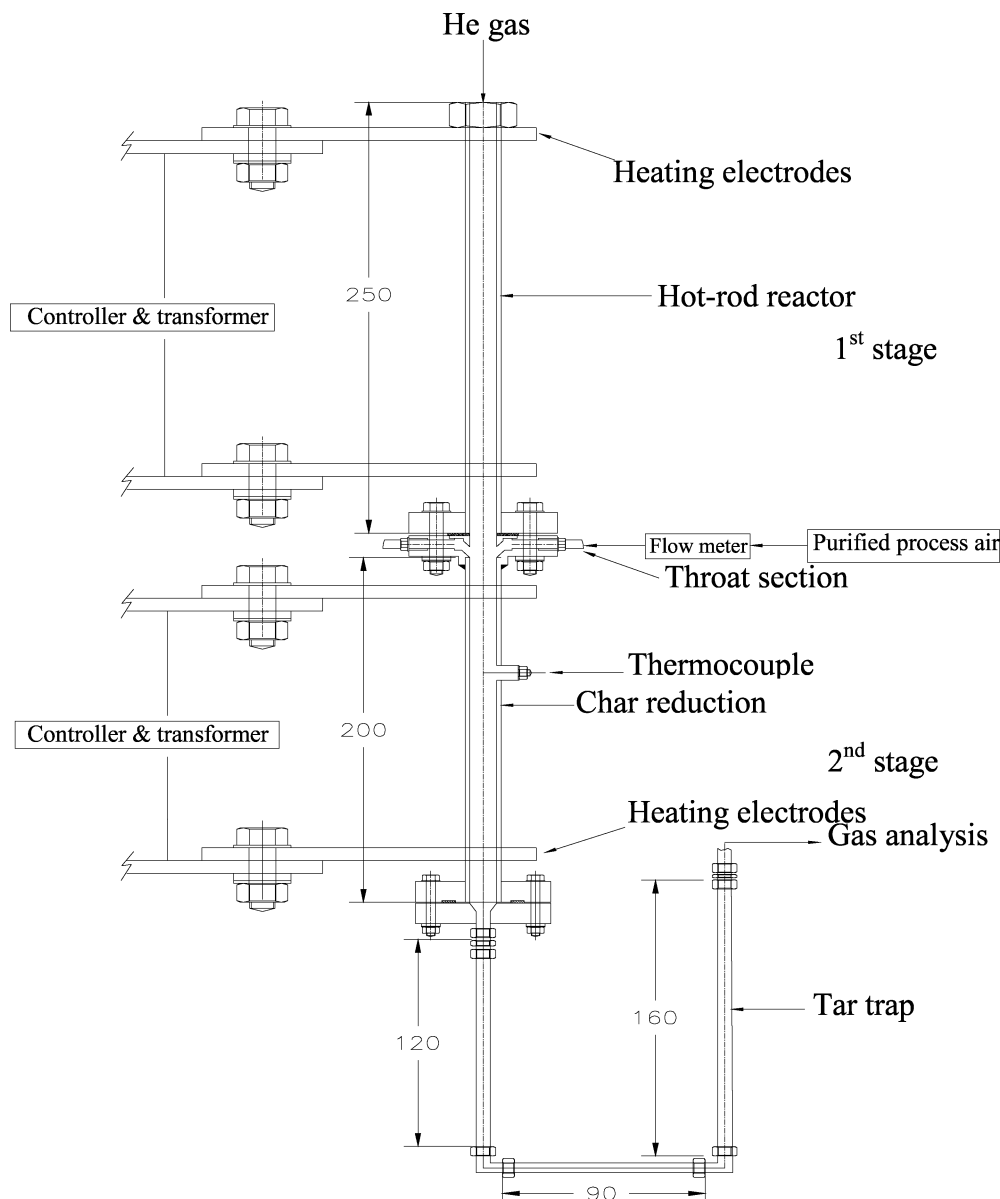
(10) Pindoria, R. V.; Chatzakis, I. N.; Lim, J.-Y.; Herod, A. A.; Dugwell, D. R.; Kandiyoti, R. Hydrolysis of sugar cane bagasse: Effect of sample configuration on bio-oil yields and structures from two bench-scale reactors. *Fuel* **1999**, *78*, 55.

(11) Gonenc, Z. S.; Bartle, K. D.; Gaines, A. F.; Kandiyoti, R. Effect of secondary reactions on molecular mass distributions of coal pyrolysis tars produced in a hot-rod reactor. *Erdoel, Erdgas, Kohle* **1990**, *2*, 82.

(12) Gonenc, Z. S.; Gibbins, J. R.; Katheklakis, I. E.; Kandiyoti, R. Comparison of coal pyrolysis product distributions from three captive sample techniques. *Fuel* **1990**, *69*, 383.

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**Figure 2.** Modified hot-rod reactor.

The new design uses the original reactor section to pyrolyze biomass under a range of conditions (temperature, biomass type, and time) and to study the effect of conditions in the second stage on the quantity of tar present in the final exit gas from the two-stage system. The aim was to identify conditions that tend to minimize the tar content of the product gas. The revised design of the hot-rod reactor to simulate a downdraft gasifier is shown in Figure 2.

In the new design, the first-stage hot-rod reactor, which represents the pyrolysis section of a downdraft gasifier, was connected to the second stage by means of a pair of flanges (Figure 3). The temperature of the first stage, where biomass was heated and volatile matter released, was limited to 500 °C, and the pressure was atmospheric. A sample weight of 1 g was used, and the heating rate to the peak temperature was 10 °C s<sup>-1</sup>. A low, downward flow rate of inert gas (helium) was used to sweep the products of pyrolysis into the throat.

The flange connection with the throat was sealed with a copper gasket. The throat was comprised of a flange with a removable

centerpiece, which contained the V-shaped throat. It contained a set of three, equi-spaced, lateral nozzles. Inert or reactive gases (e.g., diluted air) could be added via the nozzles into the zone immediately below the base of the throat.

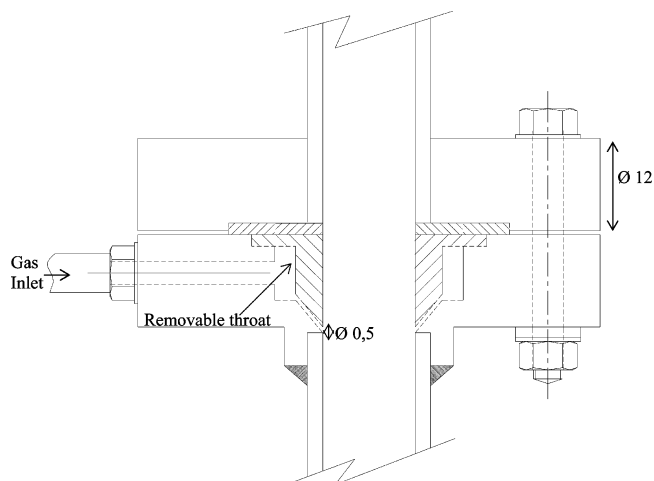
The design simulates, at the laboratory scale, the nozzles and throat of a real downdraft gasifier. The length of the flaming pyrolysis/gasification zone, where reactive gases contact the char, was near realistic: about 20 cm. In commercial scale gasifiers, the temperature of the gas in this second zone increased sharply through partial combustion reactions, so that the tars were either thermally cracked or partially (or wholly) combusted and the overall gas temperature was raised to that required for further tar cracking and gasification of char. In this laboratory scale simulation, the throat was heated externally, using heating tape.

The second reactor stage also contained a char bed and represented the high-temperature reducing zone immediately below the air nozzles of a downdraft gasifier. The construction material of this second section was Incolloy 800 HT, which is suitable for a maximum temperature of 1000 °C. It had a 12 mm i.d. and was 200 mm in length with flanges at both ends.

The temperature of this stage was controlled by one thermocouple inside the reactor; two external thermocouples helped monitor the reactor skin temperature and provide one of the safety features of

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**Figure 3.** Connecting flanges between first and second stages (lower flange contains air input nozzles).

the system. All three thermocouples were connected to a PC for control purposes and for data acquisition. Two electrodes were attached to the reactor wall of the second stage to supply the power used to resistance heat the reactor. Both electrodes were mounted flexibly, so as not to constrain the thermal expansion of the second stage during heating to temperatures up to 1100 °C. The tar trap (see Figure 2) was attached to the outlet of stage two.

**Experimental Procedures.** A preweighed wire mesh plug (formed from a strip of wire mesh) was placed inside the first stage of the reactor, approximately 70 mm above the base. A known quantity of fuel was then added, so that the sample sat on top of the plug.

A known quantity of char was added to the second-stage reactor, again supported on a preweighed wire mesh plug. The two stages were then bolted together, and the thermocouples and electrodes were assembled. The tar trap was attached below the second-stage reactor, and the lower part of the trap was immersed in liquid nitrogen. The pressure was controlled using the control valve placed after the tar trap. The required gas input rates to the top of the first stage and to the throat section were set up using rotameters and needle valves. The superficial gas velocity through the first stage was 0.1 m s<sup>-1</sup> (at 500 °C). The addition of gas through the nozzles increased the velocity in the second stage to 2 m s<sup>-1</sup>. Details of the flow system have been presented elsewhere.<sup>16</sup> The heating program, controlled by a computer, was switched on, and the heating rate and maximum temperature were entered: the test was then initiated. The first stage was operated from ambient to (usually) 500 °C at 10 °C s<sup>-1</sup> with 15 min holding. By contrast, the second stage, when used, was set at the intended temperature (between 700 and 1000 °C) and kept fixed throughout the experiment.

After the run, the reactor system was allowed to cool to room temperature, while helium gas flowed through the system to prevent oxidation of the products. Once the reactor was cooled to room temperature, the gas flow was turned off, and the reactor depressurized. The inlet piece was removed, and the tar and char were recovered to determine the yields.

**Fuel Samples.** Eucalyptus wood was used during an earlier study of high-temperature pyrolysis and gasification.<sup>17</sup> In this work, eucalyptus wood, plastic waste, and an industrial sludge were available for use. The plastic waste and industrial sludge were received from P.I.R.A. (the Paper Industry Research Association). Samples were reduced to a particle size between 106 – and 150 µm, using similar procedures. The samples were dried in a vacuum

**Table 1.** Proximate and Elemental Analyses of Biomass Samples<sup>a</sup>

element	% element			
	eucalyptus wood	sludge	plastic waste	silver birch wood
moisture content	7.5	5.9	5.9	
ash content	0.5	30.4	7.0	1.9-0.3
volatile matter	78.5	59.3	76.0	
sulfur	0.11	0.80	0.30	<0.1
carbon	45.6	30.8	45.5	52.0
hydrogen <sup>b</sup>	5.5	3.6	6.2	7.0
nitrogen	0.30	3.32	0.38	0.1
oxygen <sup>c</sup>	40.4	25.2	34.7	40.8

<sup>a</sup> Results are based on % w/w dry basis. <sup>b</sup> Corrected for moisture content.

<sup>c</sup> Calculated using determined values (by difference). Analyses performed at TES Bretby Ltd.

oven at 60 °C. The plastic waste required 24 h, the sludge 72 h. A Glen Creston grinder was then used to reduce the feedstock particles to a powder. This was sieved to collect the required size range of material. All samples were pyrolyzed in the first stage, and the tar emission was measured at the exit of this stage. Silver birch wood was used in the tests reported here with the combined first and second stages. Results obtained with the other fuels will be reported in a subsequent paper.

Difficulties were experienced with plastic waste feedstock, which would not pass through the 106 µm sieve. As a result, plastic waste was used at a size of less than 150 µm. After grinding and sieving, samples were placed again in a vacuum oven and dried at 35 °C for 16 h. The dried samples were stored under nitrogen at -20 °C. The procedure avoided sample oxidation and changes in the moisture content of the sample. Elemental analyses and ash contents of the samples are presented in Table 1.

**Char Sample.** A char derived from spruce wood was used in the second stage of the reactor. It was received from Aston University and was further heat treated in this laboratory to remove residual volatiles. The material was sieved to the desired particle size (212–424 µm) and pyrolyzed at 1000 °C under nitrogen for 100 min. The pyrolyzed sample was stored in glass bottles under nitrogen until required.

**Analytical Procedures. Collection of Tar and Char Samples.** Tar and char samples were collected after each test using the following procedure. Each stage of the reactor was washed with a 1:4 methanol/chloroform solution: this recovered all tar from the beds, reactor walls, and wire mesh strips. The tar trap was washed separately with the solvent. The solutions were each filtered through pre-weighed Whatman No.1 filter paper (42.5 mm diameter) into a pre-weighed beaker. The solvent was evaporated using a Rotovap at 80 °C, at 30–40 rpm, for 30 min. After most of the solvent had evaporated, the tar was finally dried at 35 °C for 1 h in a circulating oven, in an inert atmosphere, to complete the solvent evaporation. The tars were then weighed, and the tar yields were determined. In this part of the work, the conditions used to remove the solvent were fairly severe, and low boiling point tars and oils were also removed. The intention has been to measure the amount of more intractable high boiling tars that cause deposition and corrosion problems during utilization of the fuel gas from downdraft gasifiers.

After solvent washing, the char beds and wire mesh strips were pushed out of the reactor into pre-weighed beakers and dried at 35 °C in a circulating oven for 1 h and then weighed again. The char weight was determined by subtracting the initial weight of the mesh plug.

**Analysis of the Gases Produced in the Reactor.** PTFE bags were used to collect gas samples over sequential 3 min intervals (within a total test time of 15 min). The gas composition was then measured by gas chromatography using a Shimadzu gas chromatograph (GC) 2014 with TCD and FID detectors. The GC oven was equipped with a Carboxen –1010 Plot, 30 m × 0.53 mm i.d., 30 µm capillary column. A 0.5 mL sample was injected into the gas chromatograph. The GC operating conditions used for the analysis are shown in Table 2. A standard gas with a mixture of

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**Table 2. Operating Conditions for GC**

initial column temperature	35 °C
final column temperature	200 °C
heating rate	20 °C/min
total run time	30.25 min
injector temperature	150 °C
detector temperature	220 °C
carrier gas (He) flow rate	6.3 mL/min

hydrocarbons and other permanent gases was used to calibrate the GC to quantify the amounts present. The FID could be used to detect and measure the hydrocarbon components but not H<sub>2</sub>, N<sub>2</sub>, CO<sub>2</sub>, or CO.

The TCD could be used to measure the species not measurable with the FID, except H<sub>2</sub>. H<sub>2</sub> could not be measured with the TCD because He was used as the carrier gas, and this has a similar thermal conductivity to H<sub>2</sub>. It was found that the concentrations of all of the measurable species were low and close to the detection limits for the instrument. This was due to dilution by the high He flow through the hot-rod reactor.

## Results and Discussion

**Formation of Tars in the First Stage of the Gasifier.** The biomass/waste was progressively pyrolyzed as it was heated in the first stage of the laboratory scale reactor. This was intended to simulate the upper zone of the downdraft gasifier, where the feedstock is heated and pyrolyzed, as it moves down toward the throat region. In the laboratory simulation, the feed does not gravitate downward, but the heating from ambient to 500 °C is intended to simulate the rise in temperature seen in commercial scale systems. The heating time is approximately 500 s. In the initial set of tests, the equipment was operated without the second stage, so that the quantity of tar formed in the first stage could be determined. The tar trap was connected directly to the outlet of the first-stage reactor for this part of the work. The effect of temperature (in the range of 200–450 °C) on the quantity of tar formed from eucalyptus wood, sludge, and plastic waste has been studied, under otherwise similar reactor conditions. The tar and char yields were measured during the tests, and the results are shown in Table 3. The gas yield is calculated as the balance between the original sample weight and the weights of tar and char formed. It contains both permanent gases and low boiling point organic material not included with the tar. The total volatile yield is the sum of the tar and gas yields.

Each test was performed in duplicate, and the data in the table show that the repeatability of the test results was within  $\pm 1\%$ . The quantities of tar and gas formed increased with the temperature and also varied with the type of fuel. Both tar and gas yields decreased in the series eucalyptus wood > plastic waste > sludge. There was an accompanying increase in the char yield along this series.

The data for eucalyptus wood are shown in Figure 4. The char yields decreased rapidly over the 200–300 °C range and reached a limiting value of approximately 15% by 400 °C. There was a significant increase in tar yield up to 350 °C, but by 400 °C, it had reached a maximum value of 30%. There appeared to be little gas production at 200 °C, and the sum of the char and tar yields was close to 100%. However, above 300 °C, the gas yield was high, with half of the feedstock being converted to gas. It is noted that as defined here, the gas yield will include low boiling point organic compounds that have been lost during the solvent removal stage used to isolate the tar sample. It is clear that at up to about 200 °C, negligible pyrolysis occurs, with only limited release of some low boiling point material from the fuel, which was collected as tar. The high char yields,

**Table 3. Tar, Char, and Gas Yields from First Reactor Stage**

feedstock	T (°C)	tar yield (%) (d.a.f)	char yield (%) (d.a.f)	gas yield (%) (d.a.f) <sup>a</sup>	total volatile (%) (d.a.f) <sup>b</sup>
eucalyptus wood	200	12	86	2	14
	200	17	82	1	18
	300	29	18	53	82
	300	27	17	56	83
	350	30	13	57	87
	350	30	16	54	84
	400	30	13	57	87
	400	30	14	56	86
	450	30	15	55	85
	450	30	15	55	85
sludge	200	13	85	2	15
	200	12	86	2	14
	300	17	58	25	42
	300	15	59	26	41
	350	16	58	26	42
	350	16	58	26	42
	400	19	54	27	46
	400	19	53	28	47
	450	19	50	31	50
	450	19	51	30	49
plastic waste	200	4	97	0	3
	200	7	93	0	7
	300	17	62	21	38
	300	18	67	15	33
	400	25	45	30	55
	400	25	45	30	55
	450	25	26	49	74
	450	25	25	50	75

<sup>a</sup> Calculated from char and tar yields. <sup>b</sup> Tar plus gas yield.

measured at the low temperatures in the range studied, would have been mostly relatively unreacted feedstock. The patterns of release seen with the sludge and plastic waste were similar to those of the eucalyptus, although the concentrations of tar, gas, and char varied with the feedstock.

**Commissioning of the Two-Stage Reactor.** Silver birch wood has been used as the biomass in the first stage for all of the commissioning trials with the two-stage reactor.

**First Stage.** A series of runs was repeated under the same experimental conditions to determine the amount of tar released by the pyrolysis of the silver birch wood powder at 500 °C. This is the temperature identified in the earlier tests, which gave complete release of volatiles from the fuel. The results are shown in Table 4. For these initial tests, the tar trap was directly connected to the base of the first stage.

The data show good repeatability between the different tests. An average of 47% of the biomass was released as condensable tars. A total of 21% remained as char in the first stage, and the balance must have been released as gases and volatile organic compounds. Overall, 79% of the fuel was converted to volatile products at 500 °C.

**Throat and Second-Stage Reactor.** The first stage was resistance heated to 500 °C, and the second stage was held fixed at 800 or 900 °C, with the tar trap connected to the outlet of the second stage. In the initial tests, the intermediate section containing the throat was insulated but did not have independent heating. It was found that tars condensed in the throat, where they formed a hard deposit, by polymerization of the condensed material. The only way to avoid this was to heat the throat section to 800 °C, using electrical heating tape. When this was done, the throat was found to be clean after each test.

The extent of tar destruction in the second stage was determined by two factors: the temperature of the reactor and the residence time in contact with the char bed. To assess the impact of the hot reactor, tests were performed with an empty heated second stage. The results are shown in Table 5.

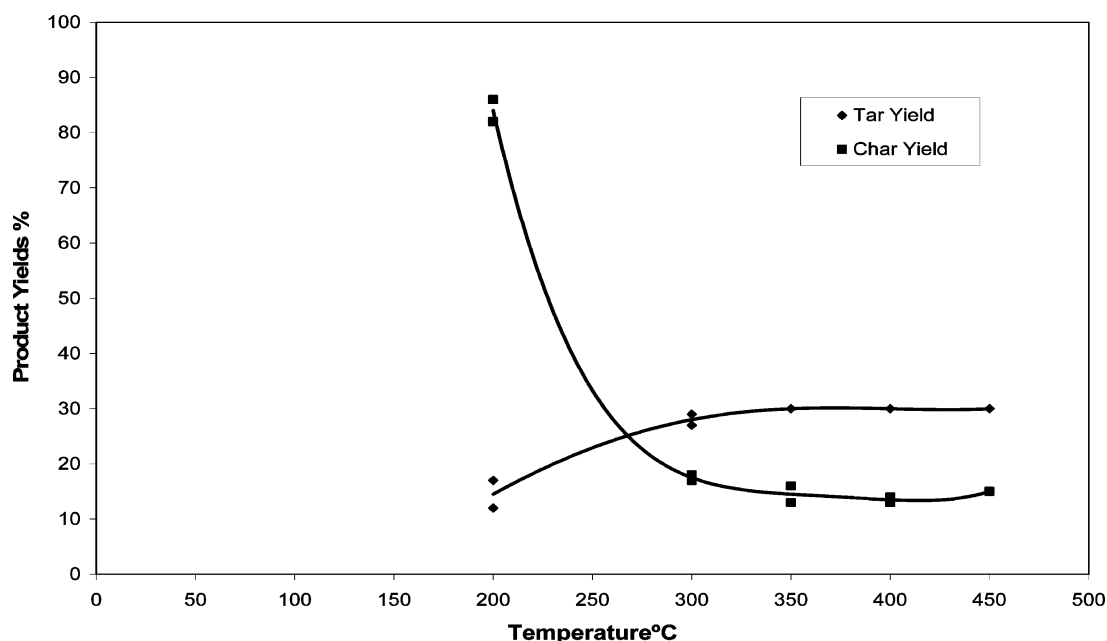


Figure 4. Tar and char yields with eucalyptus wood.

Table 4. Tar, Char, and Gas Yields from the First Stage<sup>a</sup>

tar, as % initial fuel	char, as % initial fuel	gas yield (%), calcd by difference
48	22	30
47	21	32
50	20	30
48	20	32
45	22	33
47	20	31
47	20	33
47	21	32

<sup>a</sup> Temperature: 500 °C; heating rate: 1 °C s<sup>-1</sup>; weight initial bed: 1 g (silver birch wood); and hold time: 900 s. Data in italics are the average values.

Table 5. Tar Concentration at Outlet of Heated Empty Second Stage<sup>a</sup>

<i>T</i> of second stage (°C)	tar yield after second stage (%) (by wt of initial fuel in first stage)	char % remaining in the first stage (by wt of initial fuel in first stage)
800	5.6	20
	4.9	20
	5.1	21
	5.5	20

<sup>a</sup> Temperature first stage: 500 °C; superficial velocity first stage (using He): 0.1 m s<sup>-1</sup>; gas residence time in second stage: 0.06 s

The presence of the heated empty second stage was thus observed to cut the amount of tar surviving the reactor from 47% (with no second stage) to 5.3% (average empty second stage). This shows the impact of time and temperature on the secondary reactions that progressively destroy the tar. However, it has not caused complete destruction

Table 6 shows data obtained using the two-stage configuration, with the throat heated and the second-stage reactor packed with char derived from spruce wood (particle size of 212–425 μm).

The data show that, at each temperature, the repeatability of the test data was good. Extractions carried out after each experiment showed that there was no extractable material or residual tars left in the first stage. Small amounts of tar were recovered by extraction of the char from the second stage and

Table 6. Tar and Char Yields from Two-Stage Reactor<sup>a</sup>

<i>T</i> , second stage (°C)	tar, first stage (%)	tar, second stage (%)	tar, tar trap (%)	tar, total (%)	char, first stage (%)	char, second stage (%)
800	0	0.31	0.65	1.0	21	85
	0	0.65	0.67	1.3	23	86
	0	0.51	0.64	1.2	21	84
	0	0.46	0.64	1.1	22	85
	0	0.64	0.65	1.2	21	85
	0	0.51	0.65	1.2	22	85
	0	0.51	0.65	1.2	22	85
900	0	0.17	0.20	0.4	22	85
	0	0.20	0.20	0.4	20	86
	0	0.19	0.20	0.4	21	86

<sup>a</sup> First stage temperature: 500 °C; heating rate: 1 °C s<sup>-1</sup>; weight initial bed: 1 g; and hold time: 900 s. Data in italics are the average values.

Table 7. Tar and Char Yields from Two-Stage Reactor, Using Preheated Char in Second Stage

tar, first stage (%)	tar, second stage (%)	tar, tar trap (%)	tar, total (%)	char, first stage (%)	char, second stage (%)
0	0	0.04	0.04	21	100
0	0	0.05	0.05	21	100

from the contents of the tar trap. When the second stage was set at 800 °C, this amounted to an average value of 1.2% of the initial weight of biomass in the first stage. This compares with a tar value of 47% at the exit of the first stage (Table 4). With the second stage set at 900 °C, the total tar recovery had decreased to 0.4% of the biomass charge.

The hot char bed in the second stage was thus observed to be effective in destroying tar passing through the second-stage packed bed. The data show that the weight loss of the sample fuel solids in the first stage was close to 78% (i.e., 22% remaining as char). This is consistent with the complete loss of the volatile material from the silver birch wood, as it is similar to the volatile matter content determined by the standard volatile matter determination for coals. The actual value determined by the volatile matter determination was 79%.

However, it was also observed that the packing material used in the second stage suffered a 14–15% (average) weight loss during these tests. This must be due to the presence of residual

**Table 8. Analysis of Gases Produced in Reactor<sup>a</sup>**

reactor configuration	total tar (%) fuel in stage 1	gas analysis	av concn (%)	time into test (min)				
				0–3	3–6	6–9	9–12	12–15
first stage only	47	CO <sub>2</sub>	0.40	0.63	0.16	% vol, dry 0.48	0.35	0.39
		CO	0	0	0	0	0	0.01
		CH <sub>4</sub>	0	0	0	0	0	0
first stage and empty hot second stage	0.7	CO <sub>2</sub>	0.23, 0.08	0.30, 0.06	0.34, 0.06	0.25, 0.08	0.19, 0.14	0.05, 0.06
		CO	0.16, 0.15	0, 0	0.07, 0.09	0.66, 0.50	0.08, 0.11	0, 0.04
		CH <sub>4</sub>	0.02, 0.01	0, 0	0, 0	0.10, 0.07	0.01, 0	0, 0
first stage and hot second stage with char bed (2–3 mm particle size)	0.1	CO <sub>2</sub>	0.23, 0.25	0.37, 0.10	0.32, 0.11	0.15, 0.36	0.31, 0.48	0.01, 0.21
		CO	0.41, 0.38	0.04, 0.17	0.63, 0.15	0.97, 0.92	0.28, 0.41	0.12, 0.27
		CH <sub>4</sub>	0.06, 0.05	0, 0	0.03, 0.12	0.14, 0.09	0.10, 0.04	0.01, 0

<sup>a</sup> First stage temperature: 500 °C; second stage temperature: 1000 °C; and flow gas: He (0.3 L min<sup>-1</sup>).

volatile matter contained in the char used for packing the second stage. The test at 900 °C was repeated using char that had been preheated to the test temperature, and these data are shown in Table 7.

The total tar yield at 900 °C decreased from 0.4 (Table 5) to 0.05% by the use of the preheated char in the second stage. This shows that residual volatiles from the second-stage char represented the major part of the low tar emission from the char bed (see Table 5), operated at 900 °C. It is also observed that the weight of char in the second stage was stable, confirming that it did not contain residual volatiles at the start of the test. All further tests using char in the second stage were performed using the preheated material.

The tar released from the first stage of the reactor will have contained some water (produced from the H and O in the biomass sample). It could be argued that this could have gasified some of the char in the second stage, leading to a weight loss. This was not observed; however, it is possible that the loss could be compensated for by the deposition of C from the complete breakdown of the tars. This cannot be ruled out and could account for a portion of the unaccounted material shown in Figure 5. The deposition of this secondary C could be part of the normal process, which removes tar C from the fuel gas and would occur in a downdraft gasifier. Removal of this C is clearly beneficial in terms of tar destruction, but the deposition could reduce the overall reactivity of the char in the reducing bed of the gasifier.

Adding char to the second stage has further reduced the outlet tar concentration to less than 0.1% (Table 7). However, even this low concentration can potentially cause operating problems with a plant that will be expected to run for months on end without problems. To illustrate the potential for contamination, the tar released for a plant fed with 200 kg h<sup>-1</sup> biomass has been estimated. On the basis of the data obtained at 900 °C with preheated char in the second stage, the quantity of tar in the outlet gas would be nearly 2.5 kg day<sup>-1</sup>, and over a 6 month period of continuous operation, this accumulates to nearly 0.5 tonne. This could accumulate in pipework, filters, and valves. It could also cause erosion and corrosion in the power generator (e.g., a reciprocating engine). There is thus scope for optimizing the conditions in the second stage further, to reduce the tar content, with the aim of complete destruction: parameters for study include temperature, residence time, nature of bed material, and particle size.

**Analysis of Gases Produced in the Reactor.** The analyses of the gas samples collected during tests with (1) the first stage only, (2) the first and second stages, connected and heated but without char, and (3) with the first and second stages connected, heated, and with a char bed are shown in Table 8. The table shows the total amount of tar produced, the average gas analysis, and the analyses of the individual gas bags. The only gaseous

**Table 9. Quantities of Tar, CO, and CH<sub>4</sub> at Exit of Second Stage**

reactor configuration	total tar, % (mg)	gas constituent, L, (mg of C)	
		CO	CH <sub>4</sub>
first stage only	47 (470)	0	0
first stage, plus empty heated second stage	5.3 (53)	0.15 (76)	0.02 (10)
first stage, plus heated second stage with char	0.05 (1)	0.31 (157)	0.04 (20)

species detected were CO<sub>2</sub>, CO, and CH<sub>4</sub>. H<sub>2</sub> could not be detected using the GC configuration available for this work.

The percent concentrations were low because of the high dilution by He flow from stage 1 and the throat nozzles, which were used as the sweep gas. This is a fundamental difficulty with this reactor system, where high carrier gas velocities were used to try to suppress (as much as possible) the secondary tar cracking in the first-stage reactor.

During each of these tests, silver birch wood was pyrolyzed at a temperature of 500 °C in the first stage. We have already seen that, without a second stage, 47% of the fuel was recovered as tar. Furthermore, only a low concentration of CO<sub>2</sub> was detected in the exit gas, with almost no CO or CH<sub>4</sub>. This finding suggests that the temperature in the first stage was sufficiently low and that the carrier gas flow rate was sufficiently high for significant extents of tar cracking not to take place. Addition of the heated, but empty, second stage resulted in a substantial reduction in the tar concentration, and the detection of CO and CH<sub>4</sub>, as well as CO<sub>2</sub> in the fuel gas.

The data show that the concentrations peaked in the third gas sample bag (collected between 6 and 9 min). It is clear that these species must have been formed by the thermal breakdown of the tar released in the first stage. The reduction of products in the sample from the subsequent gas bag indicated that emissions were nearing completion after approximately 12 min and that the tar emission from stage 1 was complete by this time.

The tar emission decreased further to 0.05% when char was present in the heated second stage. This was accompanied by a further rise in the CO and CH<sub>4</sub> emissions. The increase was significant in relation to the modest decrease in the tar yield. This suggests that CO and CH<sub>4</sub> were also formed in the char bed, by the destruction of other nondetected material. The most likely candidates are VOC-type material, which would be collected in the cold tar trap but would be lost during the tar evaporation stage.

Table 9 shows the total volumes of CO and CH<sub>4</sub> in the fuel gas during tests with (1) the first stage only, (2) the first stage plus a heated empty second stage, and (3) the first stage plus a heated second stage packed with char. The volumes have been estimated from the average gas analysis (% vol) and the total



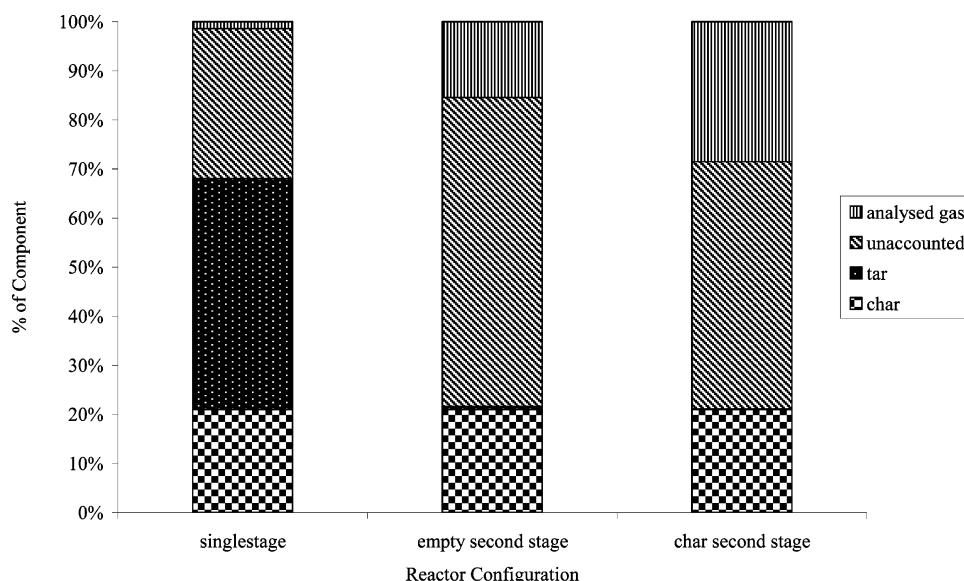


Figure 5. Distribution of biomass sample between different products.

volume flow rate of exit gas. The figure in parentheses is the number of milligrams of C present in the estimated gas volume of each component. The data show the appearance of CO and CH<sub>4</sub> when the heated empty second stage was added. The amounts of C present as CO and CH<sub>4</sub> are much lower than the decrease in the amount of tar present (C as CO and CH<sub>4</sub> is 86 mg, as compared to a tar decrease of 417 mg). When char was added to the second stage, the amount of C as CO and CH<sub>4</sub> increased to 177 mg, and the tar decreased by a further 52 mg.

The observation that the decrease in recovered tar, when char was added to the second stage, was far lower than the extra amount of C appearing in the gas as CO and CH<sub>4</sub> suggests that other species are present, which were not recovered by the present procedure for gas capture and analysis. The presence of this material is also clearly shown in Figure 5.

Figure 5 shows how the input sample (1000 mg) of silver birch wood was distributed among the measured products, with the different configurations of the reactor. The values are shown as the percent of the initial weight of the biomass. The unaccounted material is the difference between the amount of initial biomass and that measured in the products as char, tar, and permanent gases CO<sub>2</sub>, CO, and CH<sub>4</sub>.

The material unaccounted for by the present product recovery train probably represents the proportion of volatile organic species that are formed (i.e., the material intermediate between the tar and the permanent gases, —which is lost during the tar drying stage at 35 °C in the drying oven). It shows that for each configuration, there is a considerable proportion of a lower boiling point (as compared to recovered tar) volatile material and that the amount varies with the configuration of the equipment.

In the downdraft gasifier simulation, approximately 47% of the initial silver birch wood was converted to condensable organics. The amount varies with the nature of the biomass in the first stage. It is noted that in these tests, the residence time of the gas in the second-stage char bed was approximately 0.08 s. It seems that a low residence time at 800 °C is sufficient to radically alter the composition and to virtually destroy the tar, but it breaks down to lower boiling point organic material and

some gas. The destruction of the low boiling point aromatics would require a longer time at that temperature in the packed second-stage char bed.

### Conclusion

A laboratory scale fixed-bed reactor was constructed to simulate the conditions in a downdraft gasifier, and an operating protocol was developed. The performance of the equipment was validated using several biomass/waste materials. The aim of the current study was the complete destruction of tars, released from the fuel, so that commercial scale downdraft gasification equipment can operate for extended periods of time, without incurring problems caused by low residual tar emissions. This part of the study has shown that both the presence and the temperature of the char bed in the second stage have a substantial influence on the tar content of the product gas.

In these preliminary experiments, the tar content decreased from an initial value of 47% (at the outlet of the first stage) to less than 0.1% (with a hot second stage, packed with char). No combustible gases were produced from the initial pyrolysis of the biomass at 500 °C. However, the presence of the hot empty second stage and then the presence of packed char in the second stage causes a progressive increase in the amount of combustible gas through secondary cracking of the tar product. It is also apparent that some of the initial mass of the material cannot be recovered when using the tar drying procedure, described previously.

There is thus scope for optimizing the conditions in the second stage further, to reduce the tar content, with the aim of complete destruction: parameters for study include temperature, residence time, nature of bed material, and particle size. The effect of these parameters will be presented in a future study, together with an investigation of the lost condensable material.

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