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Title	Bubbling fluidised bed gasification of wheat straw-gasifier performance using mullite as bed material
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Publication date	2015-05
Publication information	Mac an Bhaird, Seán T., Phil Hemmingway, Eilín Walsh, Kevin McDonnell, and et al. "Bubbling Fluidised Bed Gasification of Wheat Straw-Gasifier Performance Using Mullite as Bed Material" 97 (May, 2015).
Publisher	Elsevier
Item record/more information	http://hdl.handle.net/10197/6471
Publisher's statement	This is the author's version of a work that was accepted for publication in Chemical Engineering Research and Design. Changes resulting from the publishing process, such as peer review, editing, corrections, structural formatting, and other quality control mechanisms may not be reflected in this document. Changes may have been made to this work since it was submitted for publication. A definitive version was subsequently published in Chemical Engineering Research and Design (VOL 97 (2015)) DOI: http://dx.doi.org/10.1016/j.cherd.2015.03.010
Publisher's version (DOI)	10.1016/j.cherd.2015.03.010

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Bubbling fluidised bed gasification of wheat straw – gasifier performance using mullite as bed material

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Abstract

The adoption of wheat straw as a fuel for gasification processes has been hindered due to a lack of experience and its propensity to cause bed agglomeration in fluidised bed gasifiers. In this study wheat straw was gasified in a small scale, air blown bubbling fluidised bed using mullite as bed material. The gasifier was successfully operated and isothermal bed conditions maintained at temperatures up to 750 °C. Below this temperature, the gasifier was operated at equivalence ratios from 0.1 to 0.26. The maximum lower heating value of the producer gas was approximately 3.6 MJm⁻³ at standard temperature and pressure (STP) conditions and was obtained at an equivalence ratio of 0.165. In general, a producer gas with a lower heating value of approximately 3 MJm⁻³ at STP could be obtained across the entire range of equivalence ratios operated. The lower heating value tended to fluctuate, however, and it was considered more appropriate for use in heat applications than as a fuel for internal combustion engines. The concentration of combustibles in the producer gas was lower than that obtained from the gasification of wheat straw in a dual distributor type gasifier and a circulating fluidised bed. These differences were associated with reactor design and, in the case of the circulating fluidised bed, with higher temperatures. Equilibrium modelling at adiabatic conditions, which provides the maximum performance of the system, showed that the gasifier was operating at suboptimal equivalence ratios to achieve greatest efficiencies. The maximum calculated theoretical cold gas efficiency of 73%

was obtained at an equivalence ratio of 0.35.

Keywords: Bubbling fluidised bed, Gasification, Wheat straw, Mullite, Gas composition, Heating value

Introduction

The Irish farming sector produces significant surpluses of cereal straw each year and the potential exists to use this biomass in energy production: the feasible straw resource for energy generation has been estimated at 80,000 - 320,000 tonnes annually (Henriksen et al., 2006; Stahl et al., 2004; Yin, 2011). There are obstacles to this use, however, amongst which is the lack of experience in this area (Hongli et al., 2009). This study investigates the use of cereal straw, specifically wheat straw, as a fuel for gasification. Gasification is a thermochemical process that has the potential to convert solid fuel into a gas with various end uses. The quality of the gas determines its most appropriate end use which spans both energy applications and chemical synthesis. The firing of the gas in boilers or its use in heat applications has been identified as the simplest end use (Maniatis, 2001).

Air, oxygen, steam, carbon dioxide, and hydrogen can all be used as gasification agents (Ren et al., 2010; Vigouroux, 2001). When using air or oxygen as the gasification agent, a portion of the feedstock is combusted to drive the endothermic gasification reactions. The portion of feedstock that undergoes complete combustion is dependent on the equivalence ratio (ER) which is the ratio of the actual air-fuel ratio to that required for complete combustion. The ER is a key parameter affecting the heating value of gas produced by gasification, and in many cases is the dominant operating parameter (de Jong et al., 2003; van der Drift et al., 2001). Specific gas yields are reported to increase continuously with ER (Mansaray et al., 1999; Natarajan et al., 1998; Sheth and Babu, 2009; Zainal et al., 2002). Generally, at low ERs the heating value of the gas increases with ER before peaking and then starting to decrease (Mansaray et al., 1999; Sheth and Babu, 2009; Zainal et al., 2002). This decrease is associated with a reduction in combustible gases and dilution with carbon dioxide and nitrogen.

Straw gasification has been shown to cause operational difficulties, for example, the low bulk density of straw makes it unsuitable for gasification in moving bed gasifiers (Maniatis, 2001). Fluidised bed gasifiers can accommodate straw which has not first been densified,

however its gasification in fluidised beds has been reported to cause severe ash sintering and bed agglomeration (Maniatis, 2001). Gasification temperature has been identified as the most important parameter in relation to the propensity of a bed to agglomerate, with higher temperatures increasing the risk (Natarajan et al., 1998). In autothermal gasification, where the heat required by the net endothermic gasification reactions is delivered by combusting a portion of the fuel, the temperature is controlled by the ER: the greater the ER, the greater the temperature. Ideal and theoretical gasification has been identified to occur at ERs between 0.19 and 0.43 (Zainal et al., 2002), thus by limiting the ER to avoid bed agglomeration the performance of the gasifier may be affected.

This study investigates the gasification of wheat straw in a small scale, air blown bubbling fluidised bed (BFB) using mullite, an alumina sand that has been reported to increase agglomeration temperatures (Ergudenler and Ghaly, 1993; van der Drift and Olsen, 1999), as bed material. The most appropriate use of the gas produced will then be determined based on the higher heating value.

Materials and methods

Apparatus and operating conditions

A bubbling fluidised bed gasifier developed by Texas A&M University (Parnell Jr. and LePori, 1988) was used to study the gasification characteristics of wheat straw. The gasifier is a 1-foot diameter, skid-mounted fluidised bed gasifier with a feed rate rating of approximately 1.8 tonnes day⁻¹ (150 lb h⁻¹) (Capareda and Maglinao, 2009). A schematic depiction of the gasifier is provided in Fig. 1 (after Capareda and Maglinao, 2009).

The gasifier was designed to accommodate cotton gin trash and similar biomass without the need for pre-treatment. A range of feedstocks have been successfully gasified to date using the gasifier including poultry litter, wood chips, cotton gin trash, dairy manure, sorghum, and switchgrass (Capareda and Maglinao, 2009; Maglinao, 2009; Maglinao and Capareda, 2008; Parnell Jr. and LePori, 1988). The gasification agent utilised in all studies was air. The design power output of the gasifier was set at approximately 73 kW based on the heating value of the producer gas from the gasification of cotton gin trash.

Fig. 1: Schematic of fluidised bed gasifier (after Capareda and Maglinao, 2009)

A natural gas burner is employed to preheat the air to the gasifier and achieve operating temperatures. The fuel is fed into the gasifier via an auger system; an optical counter is used to measure the rotational speed of the screw conveyor system feeding the fuel directly into the bottom of the bed close to the distribution plate. This feed configuration minimises segregation of the fuel from the bed which has a detrimental effect on the performance of a gasifier: segregation is linked to higher levels of tar, reduced carbon conversion efficiency, and potentially increased agglomeration (Narváez et al., 1996; Salour et al., 1993). Fuel segregation is known to occur with straw and it is recommended that the fuel be fed directly into the bed (Salour et al., 1993).

The producer gas exiting the gasifier passes through two cyclones in series: the first cyclone is designed to reduce the particulate loading to a maximum of 3 gm^{-3} and the second to a maximum 0.5 gm^{-3} . The maximum pressure drop across the two cyclones is 203 mm of water. Collection bins for the cut particles are located at the bottom of each cyclone. The turbulent, fluidised state of inert particles in the bed creates a near isothermal zone and enables accurate control of reaction temperatures (Capareda and Maglinao, 2009). To record reaction temperatures, a K-type thermocouple (Omega CAIN-14U: Omega Engineering Inc., Stamford, CT, USA) is located just below the bed base with three further

thermocouples placed along the height of the bed at 152.4, 254, and 469.4 mm above the bed base. Readings from these thermocouples are referenced as T1-T4, respectively. Pressure readings are taken at the base and the upper base of the fluidised bed by differential pressure transmitters (Omega PX274: Omega Engineering Inc., Stamford, CT, USA, and Dwyer Series 677: Northeast Controls Inc., Upper Saddle River, NJ, USA). Pressure and temperatures are continuously monitored and logged using a data logger (Omega OM-320: Omega Engineering Inc., Stamford, CT, USA).

A laminar flow element is located between the blower and the gasifier to determine the primary air flow rate to the gasifier. An online gas analyser (HORIBA Scientific, Irvine, CA, USA) located after the second cyclone was used to provide dynamic measurements of the composition of the producer gas in terms of total methane, carbon monoxide, carbon dioxide, and hydrogen. Gas samples were collected after the second cyclone in 1 litre Tedlar bags for gas chromatography analysis (Model 310: SRI Instruments, Torrance, CA, USA). For gas collection a small diameter pipe system which extended horizontally from the main gas flow was used to channel a proportion of the gas to the online analyser/Tedlar bag. The lower heating value (LHV) of the producer gas was calculated from the gas compositional readings.

Equilibrium modelling was conducted to determine the maximum theoretical efficiency of the system (Channiwala and Parikh, 2002; Mahishi and Goswami, 2007; Prins et al., 2007). Equilibrium modelling results were compiled using HSC Chemistry[®] 6.1 chemical reaction and equilibrium software (Outotec Research Oy, Pori, Finland). The modelling procedure is based on the principle of Gibbs free energy minimisation to estimate the expected chemical composition of the producer gas yield from user defined species. The gasification model adopted the fuel characteristics and operating conditions of the gasifier and accounted for the following output species: solid carbon (C), carbon monoxide (CO), carbon dioxide (CO₂), hydrogen (H₂), water (H₂O), oxygen (O₂), nitrogen (N₂), argon (Ar), methane (CH₄), ethylene (C₂H₄), and ethane (C₂H₆). The cold gas efficiency (CGE) of the gasifier during each of the tests was calculated according to the energy content of the producer gas and the energy content of the wheat straw fuel, as described by van der Drift et al. (2001).

Feedstock and bed material

Wheat straw sourced in Texas, United States of America, was utilised for all experiments.

Proximate and ultimate analyses of the fuel were conducted in accordance with Maglinao and Capareda (2008). To allow for ready fuel feeding into the gasifier, a hammer mill was used to reduce the size of the fuel to enhance feeding. Resource constraints did not allow for size characterisation of the wheat straw fuel to be conducted, however LePori and Soltes (1985) reported that variations in composition and particle size can be overcome by the violent agitation of solids in a fluidised bed gasifier, which provides efficient conversion reactions.

Mullite, an alumina sand that has been used to counteract agglomeration tendencies in fluidised beds (Ergudenler and Ghaly, 1993), was used as bed material. Its adoption has seen the successful gasification of wheat straw in a BFB gasifier of novel dual distributor type design at temperatures greater than 900 °C (Ergudenler and Ghaly, 1993). The mullite was passed through a 425 µm USA standard sieve meeting ASTM E 11 specification and the gasifier was filled to just below the disengagement zone, equal to 40 kg of bed material added to the gasifier. The purity and composition of the mullite are shown in Table 1 and the particle size distribution of the mullite is shown in Table 2. Fresh bed material was used for each gasification test. In total, three gasification tests were conducted; the average bed temperatures and ERs are detailed in Table 3.

Table 1: Purity and composition of mullite used as bed base (after C-E Minerals, 2013)

Mulcoa 47	
Al ₂ O ₃	46.8 (min 46.0)
SiO ₂	50.0
TiO ₂	1.89
Fe ₂ O ₃	0.95 (max 1.0)
CaO	0.04
MgO	0.08
Na ₂ O	0.09
K ₂ O	0.09
P ₂ O ₅	0.09
Mineralogy	
%Mullite	65
%Glass	20
%Cristabolite	15

Procedures

Gasification commenced by fluidising the mullite bed by gradually increasing the volume flow rate of air entering the gasifier until a decrease in total pressure drop followed by its

characteristic flattening out was evident. Fluidisation was achieved at superficial gas velocity of 0.27 ms^{-1} and was maintained throughout the experiment. At this point, the bed was fully fluidised and bubbling vigorously. The air was preheated with a natural gas burner to increase the reactor temperature to 500°C . After reaching 500°C , the natural gas burner was turned off and fuel (wheat straw) was introduced into the reactor. Fuel was then used to further heat up the reactor until the desired operating temperature was reached.

Table 2: Particle size distribution of mullite used as bed material.

USS Sieve	8	12	14	20	30	40	50	70	100	140	200	270	325	PAN ^a
Grade	2.36 mm	1.70 mm	1.40 mm	850 µm	600 µm	425 µm	300 µm	212 µm	150 µm	106 µm	75 µm	53 µm	45 µm	
10x18 ⁽¹⁾	0-3	10-25		60-82	0-15									0-5
10x28 ⁽¹⁾	0	15-25	45-65	6-16	5-15	0-6								0-2
14x28 ⁽¹⁾		0	1 max	30-55	35-45	10-25	5 max							1.5 max
16x30 ^(1,2)			0-3	65-75		4 max								1 max
22S ^(1,2)			TR	15-25	32-47	27-37	4-10						3 max	TR
35S ^(1,2)			TR	1-5	21-38	40-54		9-19		2-8				3 max
50S ^(1,2)				0	1-9	22-37	26-40	12-22	6-16	1-6				3 max
60S ^(1,2)					0	0-5	30-48	30-44	9-22	2-7				3 max
20x50 ^(1,2)			TR	0-8	20-50		50-72						2 max	TR
25x80 ^(1,2)				0-5	20 min 30 avg		80-93	7-12						3 max
50x100 ⁽¹⁾						TR	5-20		70-86	0-15	3 max		1 max	TR
60x200 ⁽¹⁾							0	0-11		65-90	5-20	0-6		3 max
200 IC-C ^(1,2,3)									TR		15-25			75-85
325 IC-C ^(1,2,3)									TR				5-15	85-95

Grade available in AL₂O₃ content (1) 47% (2) 60% (3) 70%

^aPAN designates the percentage of material passing the last reported screen for each size.

Table 3: Average operating conditions adopted in the three gasification tests investigating the gasification of wheat straw in a small scale, air blown bubbling fluidised bed gasifier.

Test	Fuel	Average equivalence ratio	Average bed temperature (°C) ^a
1	Wheat straw	0.27	770
2	Wheat straw	0.23	728
3	Wheat straw	0.17	654

^aThe bed temperature is taken as the average of the readings from thermocouples T2, T3, and T4

After fuel feeding had commenced and the gas burner had been turned off, the fuel feed rate was steadily decreased to obtain the desired operating conditions of temperature and equivalence ratio. The desired air-to-fuel ratio was obtained by adjusting the speed of the screw conveyor auger of the feeding system and the air flow (Maglinao and Capareda, 2010). For the first two tests the target temperature was 700 °C. For the third test the target temperature was lower to avoid loss of isothermal conditions. In this test, the fuel feed was initiated when the target temperature of 620 °C was reached and the fuel was fed at an initial ER of approximately 0.35 before being reduced to 0.15 over a short period of time.

The design of the gasifier did not allow for ash to be removed directly from the gasifier chamber for inventory control. During operation, the level of char in the collection bins located at the bottom of each cyclone was continuously monitored and emptied when required. The char was transferred to stainless steel containers which were sealed to reduce air exposure and the potential oxidation of the hot char.

The composition of the gas was measured in Tests 1 and 3 by the online gas analyser. The online gas analyser was calibrated utilising standard gases and ambient air before each test. For offline gas analysis conducted in Test 2, the gas chromatograph was calibrated utilising standard gases. Once a gas sample had been collected, the composition analysis was conducted in triplicate (n=3) as soon as possible thereafter to minimise any gas egress. The gas composition was then compared to that from the online gas analyser used in Tests 1 and 3 to further ensure the accuracy of the calibration of the online gas analyser. Following each gasification test, and once the gasifier had cooled, the interior of the reactor was inspected and the bed material removed via vacuum; any material that had adhered to the walls of the reactor was removed with a wire brush. The reactor was subsequently filled with fresh bed material for the next test.

Heating value and end use of the gas

The composition of the gas from each test was used to determine the higher heating value of the produced gas, and from this the lower heating value was calculated. The most appropriate use of the gas (i.e. in energy applications or chemical synthesis) was then determined.

Results and discussion

Gasifier operation

Figs. 2 and 3 show the temperature and pressure readings recorded during the three gasification tests.

Fig. 2: Temperature readings within the mullite bed of a small scale, air blown bubbling fluidised bed gasifier when gasifying wheat straw during (a) Test 1; (b) Test 2; and (c) Test 3. Temperature readings T2, T3, and T4 from thermocouples positioned 152.4 mm, 254, mm and 469.4 mm above the bed base, respectively.

Fig. 3: Pressure readings from within the air blown bubbling fluidised bed gasifier during gasification of wheat straw during (a) Test 1; (b) Test 2; and (c) Test 3.

As stated earlier, higher gasification temperatures were achieved in Tests 1 and 2 than in Test 3. In Tests 1 and 2, the dynamics of the bed were observed to be affected when temperatures rose above 750 °C: at approximately 750 °C, the temperatures along the height of the bed were seen to align before the temperature of the upper bed rose and the highest temperature was recorded there (note the significant change in temperature recorded by T4 thermocouple in Fig. 2a), indicating stratified combustion of biomass above the bed and poor bed fluidisation. This was followed by a pronounced decrease in the

pressure differential across the bed, which occurred 2,299 s after the upper bed became the hottest region of the bed in Test 1. In Test 2, a similar pronounced pressure drop was evident much earlier, at 830 s after the temperature in the upper bed rose, though not as substantially as in Test 1 (Fig. 2b). These temperature and pressure excursions suggest channelling within the bed, allowing fuel to pass through the bed and combust in the vapour space above it (Scala and Chirone, 2008). The data indicate that changes in temperature alignments within the bed point to the onset of defluidisation at approximately 750 °C, beyond which a detrimental effect on the dynamics of the bed was evident; this temperature closely corresponds to the 760 °C reported by Salour et al. (1993) during gasification of wood and rice straw.

For Test 3, during which the gasifier was operated at a lower temperature of 620 °C, the temperatures within the bed did not exceed 683 °C and reduced with height above the bed. The temperatures along the height of the bed were seen to track one another: the average difference in temperature readings from T2 and T4 remained less than 3.5%. Such isothermal conditions are a known advantage of fluidised bed gasifiers over fixed bed designs (Bridgwater, 2003) and demonstrate their superior heat and material transfer characteristics between gaseous and solid phases (Warnecke, 2000). In Test 3, no large sustained or sudden variations in pressures within the gasifier were evident. These temperature and pressure readings indicate stable gasification conditions.

The gasifier was operated at higher ERs in Tests 1 and 2 (0.27 and 0.23, respectively) than in Test 3 (0.17). The average ER of Test 3 includes the initial heating period where fuel was fed at a relatively high ER for 492 s in order to achieve the desired operating temperature. When this initial heating period is omitted the average ER was 0.15. This is a relatively low ER for biomass gasification; ideal and theoretical gasification has been identified to occur at ERs of between 0.19 and 0.43 (Zainal et al., 2002), and is considerably lower than the ER of 0.25 associated with maximising the mole fraction of combustibles in air gasification of wheat straw (Ergudenler and Ghaly, 1993). Up to the point of temperature convergence observed in Tests 1 and 2, the gasifier was operated at an average ER of 0.20 and 0.22, respectively.

Cold gas efficiency

Equilibrium models represent the maximum gasification efficiency (Channiwala and Parikh, 2002; Mahishi and Goswami, 2007), however it is deemed difficult to match equilibrium performance for gasification temperatures lower than 1000 °C (Prins et al., 2007). It was seen that under equilibrium conditions there is a general increase in cold gas efficiency (CGE) with ER (a trend also reported in literature). The maximum CGE of 73% was obtained at an ER of 0.35. This point of operation coincides with the carbon boundary point, the point at which carbon is no longer produced by the system. This maximum CGE occurs at a higher ER than that at which the gasifier was operated: 0.27, 0.21, and 0.17 for Tests 1-3, respectively (Fig. 4). This indicates that the operating conditions were sub-optimal, however were deemed necessary to ensure isothermal conditions were maintained during gasification.

Fig. 4: Cold gas efficiency of modelled gasification vs the ER of gasification tests

Heating value and end use of the gas

The proximate and ultimate composition of the wheat straw gasified in the BFB gasifier is reported in Table 4.

Table 4: Proximate and ultimate composition of the wheat straw gasified in this study.

Fuel	Wheat straw
Moisture content (wt %) as received	8.02 ± 0.28
Volatile content (wt %) as received	81.03
Fixed carbon (FC) (%)	8.21

C content (%)	43.51
H content (%)	5.43
N content (%)	0.58
O content (%)	39.13
S content (%)	0.12
Ash content (%)	11.24

Feedstock moisture content is known to have a substantial effect on the efficiency of gasification which can be either positive or negative depending on the moisture content of the feedstock. Asadullah (2014) reported that under the temperatures achieved during gasification, the water contained in the feedstock is converted to steam, a gasification agent used to convert volatiles to producer gas. If the moisture content is excessively high, however, the efficiency of the process is reduced as the moisture absorbs a greater proportion of heat, increasing the energy requirement to convert the biomass into producer gas (Asadullah, 2014). In this case, the moisture content of the straw as received was $8.02 \pm 0.28\%$ which is considered acceptable for gasification as Kaewluan and Pipatmanomai (2011) successfully co-gasified feedstocks with moisture contents as high as 27% in a fluidised bed gasifier.

Fig. 5 illustrates the HHV of the gas produced during this experimental work and compares it with the HHV of the gas produced during the gasification of cotton gin trash using the same BFB gasifier (Craig, 1980). It is evident that the HHV of the gas produced from cotton gin trash is generally greater; at lower fuel-to-air ratios a convergence with the HHVs recorded in this experimental work occurs, however. The HHVs from this work are also noted to be lower than the 5.59 MJm^{-3} reported by Capareda and Maglinao (2009) when the BFB gasifier was fuelled with poultry litter and wood chips at a similar temperature (760°C vs. 770°C in Test 1) as well as the HHVs associated with the gasification of agricultural residues in BFBs at a similar temperature ($4.85 - 5.87 \text{ MJm}^{-3}$ at 760°C (Salour et al., 1993)).

Fig. 5: A comparison of the higher heating values of the gas obtained from the gasification of wheat straw and cotton gin trash in an air blown bubbling fluidised bed gasifier.

The gasification of wheat straw in the BFB gasifier in this study produced a maximum LHV of 3.6 MJm^{-3} at an ER of 0.165. A producer gas with LHV of approximately 3 MJm^{-3} at standard conditions of temperature and pressure (STP) was obtainable at a range of ERs. In all tests, the LHV of the gas exceeded the minimum heating value required for use in internal combustion engines, reported by Knoef (2005) to be 2.5 MJm^{-3} at STP. Knoef (2005) stated that a heating value in excess of 4.2 MJm^{-3} at STP is preferable; indeed, literature indicates that engines are typically run on gases meeting this more demanding limit. Although the LHV at STP justifies the use of the produced gas as a fuel for internal combustion engines, the necessary cooling of the gas to STP for use in internal combustion engines (Bridgwater, 1995; Stevens, 2001) can cause the condensation of tars which can lead to engine failure (Stevens, 2001). An average tar value of 10 gm^{-3} at STP has been reported for fluidised bed gasifiers (Milne et al., 1998). To facilitate the use of the gas obtained in these tests in internal combustion engines, secondary tar reduction measures would therefore be required to reduce the tar loading to the tolerance level of internal combustion engines, reported to range from as low as 1 mgm^{-3} at STP to 100 mgm^{-3} at STP (Bhattacharya et al., 2001; Boerrigter and Rauch, 2006; Cao et al., 2006; Han and Kim, 2008; Lettner et al., 2007).

In contrast to the use in internal combustion engines of gas obtained from the gasification of wheat straw, only particulates must be reduced for use in heat applications (Maniatis, 2001). Such applications have been identified as the simplest use of producer gas,

nonetheless there are very few examples (Maniatis, 2001). The Kymijärvi power plant in Finland, operated by Lahden Lämpövoima Oy, is an example of a gasification project where low calorific producer gas (typically between 1.6 and 2.4 MJm⁻³) has been successfully co-fired in a coal-fired boiler using a fluidised bed of circulating design to produce electricity for the owner and district heat for the city of Lahti (Raskin et al., 2001). The LHV of the gas produced in this study exceeds this range, thus heat applications are considered to be the most suitable end use for this gas rather than its use in internal combustion engines.

Conclusions

In order to maintain isothermal bed conditions, the temperature of the bed was maintained below 750 °C; under these temperature conditions the gasifier was operated at ERs from 0.1 to 0.27. Equilibrium modelling showed that the gasifier was operating at suboptimal ERs: the maximum calculated theoretical CGE of 73% was obtained at an ER of 0.35.

The maximum LHV of the producer gas was approximately 3.6 MJ m⁻³ and was obtained at an ER of 0.165, well below the modelled ER to obtain maximum CGE. It was observed that, despite fluctuations, a producer gas with an LHV of approximately 3 MJm⁻³ at STP could be obtained across the range of ERs. Considering the LHV obtained, and taking fluctuations in LHV into account as well as the reported tar content for fluidised bed gasifiers, it is suggested that the optimal use for the producer gas is in heat applications.

Acknowledgements

This study was funded under the Charles Parsons Energy Research Award of Science Foundation Ireland [Grant Number 6C/CP/E001] supported by the Department of Communications, Energy and Natural Resources of the Government of Ireland.

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