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Production of hydrogen-rich syngas from steam gasification of blend of biosolids and wood using a dual fluidised bed gasifier

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ABSTRACT

A study was undertaken on steam gasification of mixtures of wood pellets and biosolids (dried sewage sludge). The gasification experiments were conducted in a fast circulating dual fluidised bed gasifier. In the experiments, the gasification temperature was set at 720 °C and the fuel feeding rate to the gasifier was 15.5 kg/h (as received). The biosolids was blended with wood pellets, with biosolids to wood pellet mass proportion being controlled at 0% (or 100% wood pellets), 10%, 20%, 40%, 60%, 80% and 100%. From the results, the syngas produced from the biosolids had higher H₂ content (28%) compared with that from pure wood (23%), with the H₂ content increasing with biosolids fuel loadings. In addition, the ratio of H₂/ CO increased from 0.6 to 0.9 as the biosolids loading increasing from 0% to 100%. However, the syngas yield and the cold gas efficiency of the gasification decreased dramatically by 53% and 43%, respectively, at 100% biosolids loading compared with 100% pure wood loading. Nevertheless, the increase from 10% to 20% loading of biosolids in the fuel did not diminish the yields and the cold gas efficiency. For gasification of pure biosolids, the gas concentrations of H₂ and CO in this study were found to be 40% higher than that produced by other researchers using air, O₂ or CO₂/N₂ as the gasification agent.

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1. Introduction

Biosolids is defined as dried sewage sludge, which is a mixture of carbonaceous, phosphorous and nitrogenous compounds with heavy metals and (in its wet state) microbial organisms. The phosphorous and nitrogen-containing compounds make biosolids a potentially valuable fertiliser. However, the contamination of heavy metals, toxins, dioxins and microbial contamination that are present in biosolids makes it unsuitable on farmland due to the risk of the contaminants entering the food chain [1,2]. On the other hand, the high carbon content of the biosolids can be utilised as a renewable fuel. The management and disposal of biosolids, the digested and dehydrated solid product from sewage sludge, have become an important environmental issue. Major high-density population centres are increasingly looking for new environmentally benign disposal methods for the rapidly increasing biosolids. Motivating factors for sewage sludge treatment include meeting environmental legislation requirements relating to the traditional options of biosolids disposal, such as land-filling and use as agricultural fertiliser, and thereby reducing the rising cost of biosolids disposal in the landfill. As an example, in 2005 an estimated 1.7 million tonnes of dried sewage sludge was generated in the UK, with 45% being used in agriculture, 38% used in incineration and power generation, and 17% going to landfill [3]. In New Zealand, approximately 240,000 dry ton-

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nes of biosolids was produced in 2006, with approximately 50% being used for land reclamation, 35% to landfill and the rest being used in forest application and other beneficial reuse [4]. Here, land reclamation refers to conversion of unusable land into usable land, whereas landfill is an area used for controlled disposal of solid waste to generate landfill gas to produce heat and electricity [4]. Therefore, a new technology is required in order to maximise the energy recovery of the biosolids, which can potentially be a substitute for fossil fuel rather than merely for land reclamation or landfill.

The study by Rulkens [5] has identified nine options for energy recovery from biosolids. Gasification of biosolids is selected for the present study because gasification (with combined cycle) in general is a more efficient process than incineration or combustion of biosolids for power generation [5,6]. The primary product from the gasification process is a light reactive gas mixture called syngas, which consists of mostly hydrogen (H2), carbon monoxide (CO), methane (CH₄), carbon dioxide (CO₂), small amounts of nitrogen (N_2) and light hydrocarbons, and tars. Furthermore, gasification can be operated at lower temperatures to convert solid fuel to energy, with a consequential smaller quantity of greenhouse gases generated, such as nitrogen and sulphur oxides. In addition, the gasification process provides the flexibility of converting solid fuel into syngas to meet the specific syngas compositions required for downstream applications, such as fuel cells, Fischer Tropsch (FT) or dimethyl ether (DME) synthesis. Depending on the gasification medium, the syngas calorific value ranges from 4 MJ/Nm³ for air gasification to 16 MJ/Nm³ for steam gasification. The high end

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values are comparable with those from digester or landfill biogas, and the gas is suitable for combined heat and power (CHP, "cogen") applications [7]. However, the current research is focusing on potential use of the syngas as a feedstock for FT synthesis.

Recently, gasification of biosolids has attracted great interest, both in the scientific community and industry, as an alternative to disposal by incineration or combustion [8-10]. The use of biosolids as a fuel also maintains the carbon-neutrality of the process. Numerous studies published are focussed on applying gasification and pyrolysis in processing of the biosolids [2,11–16]. Co-gasification and co-firing of biosolids with coal have also been extensively investigated [17,18]. However, the main constraint of using biosolids as a fuel is the high ash content of approximately 30% by weight, which has the tendency of causing bed agglomeration and slagging in a fluidised bed both in the gasification and combustion. These issues can be mitigated by co-gasification of biosolids with low ash content biomass such as radiata pine wood, which has ash content of less than 1%. However, to date no such investigations of co-gasification of biosolids with biomass have been reported on the influence of the mixing ratio of the fuels on the gasification performance such as syngas compositions, syngas yields and the cold gas efficiency.

Formation of tars during gasification of both woody biomass and biosolids is a technical issue hindering development of biosolids gasification. Tars cause significant problems for downstream processes. The tars generally consist of several aromatic rings of organic compounds [19]. However, tar content and composition in biosolids gasification could be slightly different from woody biomass gasification due to the high ash content of biosolids, which may have a catalytic effect on tar cracking. Therefore, further studies are required to provide the information of tar concentration in the co-gasification of biosolids with woody biomass.

A dual fluidised bed steam gasifier, which consists of a combustion and a gasification reactor, is typically operated at a lower temperature (700–900 °C) than a fixed bed or an entrained flow gasifier. The ash component of the biomass or the biosolids at such high temperatures can become 'sticky' and agglomerate, resulting in defluidisation. The use of steam as a gasification medium produces higher calorific value syngas than that using air. However, steam gasification is an endothermic reaction and agent requires heat input. One way to provide the required heat is to use a combustion reactor in which the solid char produced from the gasification is combusted and the bed material is heated at the same time. The heated bed material is then circulated to the gasification reactor for heat supply. This dual fluidised bed gasification technology has been successfully demonstrated by the Vienna University of Technology in Austria [20,21].

The aims of the present study are: (i) to investigate the influence of biosolids proportion on syngas production, cold gas efficiency and syngas composition; (ii) to examine the tar concentration as a function of the biosolids proportion in the fuel; and (iii) to compare the syngas compositions of this study with previous studies which used air, O_2 and CO_2/N_2 as the gasification medium.

2. Experimental setup

The gasifier used in this study is a dual fluidised bed design, consisting of a bubbling fluidised bed (BFB) gasification reactor

fluidised by steam, and a circulating fluidised bed (CFB) combustion reactor, capable of handling a fuel feed up to 20 kg/h (approximately 100 kW_{th}). The BFB has an internal diameter of 200 mm with a height of 2 m and the CFB has an internal diameter of 100 mm with a height of 3.7 m. In the experiment, the fuel, which consisted of varying proportions of granular biosolids and woody biomass pellets, was fed via screw auger into the base of the BFB, where the gasification process occurred, forming the producer gas, termed as syngas here. This was achieved by intimate mixing of the fuel with the bed of sand particles, fluidised by the steam. In this study, Greywacke river sand was used as the bed material and its major elements are presented in Table 1. Greywacke sand was chosen for the bed material for its inert nature and lack of reported catalytic activity. The steam, at 200 °C and 6 bar (gauge), was introduced at the bottom of the BFB, where the solid char and the bed material were also transferred by differential pressure through the chute to the CFB. In the CFB, the solid char and supplementary liquefied petroleum gas (LPG), at 25–30 kW. were combusted with injected air, heating the bed material. Here, the injected air can be divided into primary and secondary air. The primary air at the bottom of the CFB was used to fluidise the solid material above the secondary air. The secondary air, which was introduced 200 mm above the primary air nozzles, was used in order for entrainment and circulation to occur. In the CFB, the bed material was fluidised upwards and carried out of the column into the cvclone that separated the heated bed material from the combustion flue gas, as shown in Fig. 1. The bed material was then delivered back to the BFB through the siphon. A schematic diagram of the dual fluidised bed gasifier is given in Fig. 1. Details of the design and construction of the gasifier are described elsewhere [22].

The biosolids used in the present study were supplied by a local waste water treatment plant and sewage sludge processor. The biosolids supplied had been biologically digested then dried in combined rotary and moving belt dryer in a separate operation and were supplied as a bulk sample in granular form with a moisture content of 8% (dry basis). Batches of premixed blends of biosolids and wood pellets were made up, with the biosolids proportion being controlled at 10%, 20%, 40%, 60% and 80% (w/w). In addition, batches of pure wood pellets and pure biosolids were also prepared for the experiments. The high heating value (HHV) and chemical compositions of biosolids and wood pellets are as shown in Table 2. The batches were fed to the gasifier through the existing feeder system consisting of a hopper and auger feeder. Approximately 5 L/min of N₂ was introduced into the hopper as a purge gas to counter the back pressure of the syngas from the BFB. The introduction of the biosolids was initially thought to possibility lead to compaction and binding problems in the auger, and so samples were taken from each batch from the test fuel to test the binding strength of the biosolids when compressed with wood pellets. However, it was found that the biosolids did not cause any binding or compaction problems of the fuel blends in the feed auger. Overall, the feeder system coped smoothly with the fuel blends. The fuel blends were fed at a rate of 15.5 kg/h (as received) equivalent wood pellets mass flow rate. The ratio of steam to fuel mixture in the present study was set at 1.1 (kg/kg_{fuel}).

In the experiments, the gasification temperature was set at 720 °C. Due to the nature of the biosolids, a large quantity of solid dust was formed and was entrained out with the syngas. Most of the particulates were separated by the cyclone prior to downstream application. It was also found that tars were

Table 1

XRF analysis result of the major elements (wt%) present in the Greywacke sand. Note that Loss on Ignition (LOI) represents the volatile components such as hydrated water, carbonate and carbon present in the Greywacke sand.

SiO ₂	Al_2O_3	Na ₂ O	Fe ₂ O ₃	K ₂ O	CaO	MgO	P ₂ O ₅	MnO	TiO ₂	LOI
71.32	14.42	4.06	3.59	2.51	1.89	1.39	0.15	0.05	0.01	1.53

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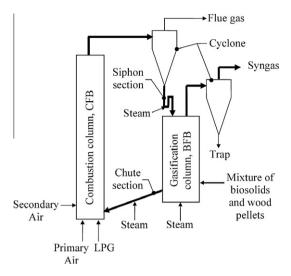


Fig. 1. Schematic diagram of the dual fluidised bed gasifier used in this study.

Table 2The high heating value (HHV) and chemical compositions of biosolids and wood pellets used in the present study.

	Biosolids	Wood pellets			
HHV (MJ/kg)	14.1	18.6			
Proximate analysis (%)					
Moisture content	8	8			
Volatile matter	43.5	77.4			
Ash content	32	0.4			
Fixed carbon	16.5	14.2			
Ultimate analysis (%)					
Carbon	34	47.2			
Nitrogen	5.1	<0.2			
Hydrogen	3.5	5.4			
Sulphur	1.2	<0.1			
Oxygen	16.2	38.7			
Ash content	32	0.4			
Moisture content	8	8			

generated from the biosolids gasification. Therefore, the produced syngas would require cleaning before any downstream application. In this case, the syngas was incinerated in an afterburner.

2.1. Syngas and tar sampling and analysis

During the gasification runs, the syngas samples were extracted in two 50 mL aliquots through a 3 mL Bakerbond amino normal phase SPE column, which trapped the tar components in the syngas for later extraction and analysis. The syngas and tar sampling system was developed by Bull [22]. The location of the sampling port was at the top of the cyclone after the BFB (Fig. 1). The first 50 mL aliquot was extracted from the syngas line using a plastic sample collection syringe in order to flush out the air trapped in the syringe; the syngas was then expelled. The second 50 mL aliquot was extracted in the same way and stored in the sample syringe, which was then sealed, removed from the sampling system and transported to an Agilent 3000 micro gas chromatography (GC) for analysis. For the tar analysis, the total tar concentration in the syngas for this study was determined by evaporative analysis adapted from Xu et al. [23]. The tar in the SPE column was eluted with 3 mL of dichloromethane (DCM) into an evaporating dish. The tar/DCM/water mixture was then heated in an oven at 105 °C for one hour to remove water and DCM in the sample. The weight of the residue (tar) was determined by the weight of the dish before the elution and after the heating. The total tar concentration in the syngas was determined by the weight of the residues over the total volume of sampled syngas (100 mL) and then converted to g/Nm³. Note that the measured tar concentration may be found to be lower than the actual tar concentration due to evaporation of Class 2 (heterocyclic compounds), Class 3 (aromatic compounds) and some losses on Class 4 (light poly-nuclear aromatic hydrocarbons) during the removal of moisture and solvent from the sample.

3. Results and discussion

The results of change in syngas composition as a function of biosolids proportion in the feed fuel are shown in Fig. 2. In the figure, each point represents an average value of three repeatable measurements. The percentage of the biosolids loading in the fuel has a significant influence on the gas compositions of CO, H₂, CO₂ and N2. However, the gas composition of CH4 and light hydrocarbons (LH) were found to be constant. The CO concentration decreased linearly from 40% to 32% as the biosolids fuel loading was increased from 0% to 100%. However, the H₂ concentration was found to be constant at 23% with the biosolids proportion varying from 0% to 20%. On the other hand, the H₂ concentration increased gradually from 23% to 28% with further increasing of the biosolids proportion from 20% to 100%. For the CO₂, the concentration increased significantly from 17% to 23% as the loading of biosolids was increased from 0% to 10%. Conversely, the CO₂ concentration gradually decreased from 23% to 10% as the loading of biosolids was further increased from 10% to 100%. As expected, the N₂ concentration increased gradually from 0.1% to 10% as the loading of biosolids was increased from 0% to 100%. The increase of N2 concentration in the syngas resulted from the increase of nitrogen content in the biosolids within the fuel mixture. Here, the N₂ concentration was calculated based on the subtraction of the N₂ concentration in the syngas with the amount N₂ fed into the hopper. The increase of H₂ and the decrease of CO and CO₂ concentrations at above 20% biosolids loading resulted from the increase of calcium oxide (CaO) content in the gasifier as biosolids loading was increased. Note that the concentration of CaO in the biosolids was measured to be approximately 16 wt% of the total ash content as shown in Table 3. The removal of CO₂ by CaO can be explained using reactions of Eqs. (1) and (2) resulting in a higher H₂ and lower CO concentration in the syngas. Also, the presence of iron (Fe) [24] and other alkali salts such as potassium (K) and sodium (Na) [24,25] in the ash (Table 3) were considered to contribute to the catalytic activity. As a result, the ratio of H₂/CO increased significantly from 0.6 to 0.9 as the biosolids loading was increased

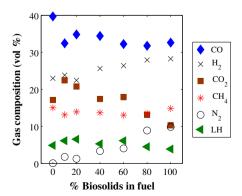


Fig. 2. The syngas composition (vol%) varying with relative proportion of biosolids in the fuel. (Note that LH represents light hydrocarbon and the error bar of the producer gases were measured to be ±15%.).

Table 3
XRF analysis result of the major elements (wt%) present in the biosolids ash.

SiO ₂	CaO	P_2O_5	Al_2O_3	Fe_2O_3	SO ₃	MgO	Na ₂ O	TiO ₂	K ₂ O	Mn_3O_4	LOI
44.47	15.63	13.57	8.82	5.19	2.41	2.32	1.88	1.6	1.55	0.12	2.44

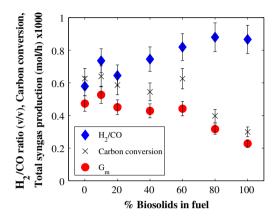


Fig. 3. The influence of biosolids proportion in the fuel on the H_2/CO ratio (v:v), the carbon conversion and the total syngas production, G_m (mol/h).

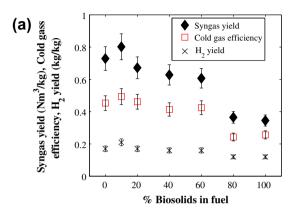
from 0% to 100% as shown in Fig. 3. However, these ratios are much lower than the optimum $\rm H_2/CO$ ratio of two for liquid fuel synthesis through FT process. Therefore, further studies on different types of catalyst as well as operation optimisation on the gasification conditions will be performed in an attempt to increase the ratio of $\rm H_2/CO$.

$$CaO(s) + CO_2(g) \leftrightarrow CaCO_3(s) \\ - 178 \; kJ/mol \tag{1}$$

$$CO(g) + H_2O(g) \leftrightarrow CO_2(g) + H_2(g) \qquad -41 \text{ kJ/mol} \tag{2} \label{eq:2}$$

The influence of biosolids proportion in the fuel on the H₂/CO ratio, the carbon conversion rate and the total syngas production. G_m (mol/h) are presented in Fig. 3. It was found that the carbon conversion rate decreased gradually from 0.62% to 0.3% for 0-100% biosolids loading. Note that the carbon conversion rate is defined as the total carbon in the syngas divided by the total carbon content found in the fuel. As a result, the total syngas production decreased from 500 to 230 mol/h. The decrease in the carbon conversion rate was due to the friable nature of the dried biosolids. There was an increase in resultant fly-ash content compared with gasification of wood pellets, and thus the fixed carbon content and the volatiles content were reduced for the biosolids. Consequently, significant quantities of biosolids entrained in the gas stream, instead of being converted into char and syngas. In a scaled-up process it is expected that biosolids preparation would involve densification, e.g. pelletising. Also, the low porosity of char/carbon and complex crystal structures of the fixed carbon in the biosolids are considered to reduce in the rate of carbon conversion. Further study on the reactivity of char of the biosolids will be conducted in order to achieve an increase in the rate of carbon conversion to increase the syngas yield.

The syngas yield, cold gas efficiency, H_2 yield and net calorific value (NCV), as a function of biosolids proportion in the fuel are presented in Fig. 4a and b. The gas volumes in Fig. 4a and b are represented as normal cubic metres (Nm³) at 0 °C and 1 atm pressure. The results are given as averaged values over three samples. Here, the cold gas efficiency is defined as the combustion energy of the syngas generated, divided by the input energy. The input energy consists of the fuel mixture (biosolids and wood pellets), LPG and steam. The syngas (Nm³/kg) and H_2 (kg/kg) yields are represented as the volumetric flow rate of the syngas (Nm³/h) and H_2 (kg/h),



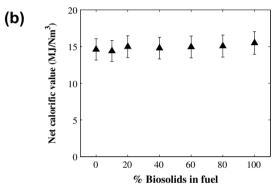


Fig. 4. The influence of biosolids loading in the fuel on: (a) syngas and hydrogen yields and cold gas efficiency, and (b) net calorific value.

respectively, which are normalised by the fuel fed (kg/h). Interestingly, for 10% loading of the biosolids in the fuel the syngas yields and the cold gas efficiency were found to be 13% and 6%, respectively, which is higher than the yields and the efficiency recorded with 100% wood pellets. However, the yields and the efficiency were found to be lower by 5% for 20% loading of biosolids in the fuel compared with that of the 100% wood pellets. A general trend can be observed in the results: with biosolids loading increasing from 10% to 100%, the syngas yield decreased by 53%, from 0.80 to $0.35 \, \text{Nm}^3/\text{kg}$ and the H_2 yield decreased by 30%, from 0.17 to 0.12 kg/kg; the cold gas efficiency decreased by 43%, from 0.45 to 0.26 (Fig. 4a). However, the NCV was found to be constant at 15 MJ/Nm³ (Fig. 4b) as the biosolids loading in the fuel was increased. The decrease of cold gas efficiency resulted from the low carbon conversion rate as described above. In part this can also be explained by the much higher ash content in the biosolids compared with wood pellets (32% compared with 0.4%). The lower syngas production was also due to the biosolids being biologically digested in the pre-processing prior to gasification, which effectively reduced some reactive carbon and volatile matters. Nevertheless, a 10-20% loading of biosolids in the fuel did not diminish the yields and the efficiency. Also, with such a low loading, the effect of defluidisation on the fluidised bed due to bed agglomeration or slagging by the high ash content of biosolids could be minimised.

The total tar concentration in the syngas, which in general, increased with increasing biosolids in the fuel except for one point

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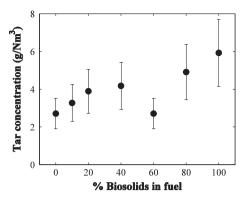


Fig. 5. The total tar concentration (g/Nm^3) in the syngas gas varying with relative proportion of biosolids in the fuel.

of 60% biosolids loading is shown in Fig. 5. In the figure, each point shows an average value of three repeatable measurements. The total tar concentration was found to increase from 2.7 to 5.9 g/Nm³ as the loading of biosolids was increased from 0% to 100%. The observation of the increase in tar concentration with biosolids loading was opposite to expectation but this may show that the syngas from the wood pellets gasification in this study has a lower tar concentration than that from the biosolids gasification. In addition, the production of volatile gas could be from higher for the biosolids than that from the wood in the initial stage of the gasification [3]. However, further studies are required in order to understand the formation of the tars from the gasification of the wood pellets, the biosolids and their blends. In biomass gasification and biosolids gasification, tar analysis is still the key area for further investigation due to the complicated composition and structure of the tars. In the future studies, a new analysing technique using a gas chromatography-flame ionised detector (GC-FID) will be explored to determine the tar concentration for each species in the tar sample. Once the tar concentration and tar species are determined, gas cleaning technologies will be investigated to reduce the tar concentration in the syngas to required levels depending on the downstream application (normally less than 1 mg/Nm³).

The comparison of syngas compositions produced from 100% biosolids in this study (steam as the gasification agent) with those reported in literature using air [2], O_2 [8] or mixture of CO_2/N_2 $(\psi CO_2 = 0.2)$ [9] as the gasification agents is shown in Fig. 6. For air gasification, Groß et al. [2] performed trials in a two-stage atmospheric pilot scale fluidised bed gasifier at an operation temperature of 870 °C. The results using O2 as the gasification agent were obtained from the study of Marrero et al. [8], who conducted the experiments in a laboratory batch scale gasifier at 1200 °C. Note that the N₂ concentration was not available in the report of Marrero et al. [8]. Petersen and Werther [9] used the gas mixture of CO₂/N₂ as the gasification agent in a pilot scale CFB operated at 800 °C. The present study using the dual fluidised bed gasifier with steam had the advantage that it was able to produce high concentrations of H₂ and CO, compared with other types of gasifier and gasification agent as mentioned above. The concentrations of H₂ and CO in the present study were found to be 40% higher than for those using other gasification agents, as shown in Fig. 6. Furthermore, the concentration of CO2 in the steam was found to be 35% lower than that using O₂ or CO₂/N₂. Therefore, the syngas from the present study using steam gasification shows a much higher NCV based on comparison with other gas composition.

4. Conclusion

The influence of biosolids loading in blended woody biomass and biosolids on fuel gasification performance has been experi-

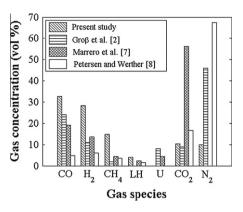


Fig. 6. Comparison of the syngas gas compositions generated from 100% biosolids using steam as the gasification agent (present study) with those using air [2], O_2 [8] or CO_2/N_2 ($\psi CO_2 = 0.2$) [9] as the gasification agent. Note that in the figure LH is light hydrocarbons and U is unidentified gas species.

mentally investigated in this study in a dual fluidised bed gasifier, using steam as the gasification agent. The key findings of this study are that the syngas produced from the gasification of pure biosolids had higher content of H₂ (28%) compared with that from pure wood (23%), with the H₂ content increasing with biosolids fuel loadings. In addition, the ratio of H₂/CO increased from 0.6 to 0.9 as the biosolids loading increasing from 0% to 100%. However, the syngas yield and the cold gas efficiency in the gasification decreased dramatically by 53% and 43% at 100% biosolids loading compared with 100% pure wood loading, respectively. Nevertheless, the addition of 10-20% loading biosolids in the fuel did not diminish the syngas and H₂ yields, and the cold gas efficiency. For gasification of pure biosolids, the gas concentrations of H₂ and CO in this study were found to be 40% higher than that produced by other researchers using air, O2 or CO2/N2 as the gasification agent. Further studies will be conducted in order to investigate the feasibility of this potential source for production of energy and liquid fuels.

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