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Exergetic evaluation of biomass gasification

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Abstract

Biomass has great potential as a clean, renewable feedstock for producing modern energy carriers. This paper focuses on the process of biomass gasification, where the synthesis gas may subsequently be used for the production of electricity, fuels and chemicals. The gasifier is one of the least-efficient unit operations in the whole biomass-to-energy technology chain and an analysis of the efficiency of the gasifier alone can substantially contribute to the efficiency improvement of this chain. The purpose of this paper is to compare different types of biofuels for their gasification efficiency and benchmark this against gasification of coal. In order to quantify the real value of the gasification process exergy-based efficiencies, defined as the ratio of chemical and physical exergy of the synthesis gas to chemical exergy of a biofuel, are proposed in this paper. Biofuels considered include various types of wood, vegetable oil, sludge, and manure. In this study, exergetic efficiencies are evaluated for an idealized gasifier in which chemical equilibrium is reached, ashes are not considered and heat losses are neglected. The gasification efficiencies are evaluated at the carbon-boundary point, where exactly enough air is added to avoid carbon formation and achieve complete gasification. The cold-gas efficiency of biofuels was found to be comparable to that of coal. It is shown that the exergy efficiencies of biofuels are lower than the corresponding energetic efficiencies. For liquid biofuels, such as sludge and manure, gasification at the optimum point is not possible, and exergy efficiency can be improved by drying the biomass using the enthalpy of synthesis gas.

Keywords: Biomass; Biofuels; Gasification; Exergy analysis; Process efficiency

1. Introduction

Biomass has great potential as a renewable and relatively clean feedstock for producing modern energy carriers, such as electricity and transportation fuels. In order to compete with fossil energy sources an optimal utilization of biomass resources is desired. For biomass-based systems, a key challenge is thus to develop efficient conversion technologies. This paper focuses on the process of biomass gasification, a technology that converts biomass into gaseous products. The gas may subsequently be used for the production of electricity, fuels and chemicals. Currently, biomass gasification is considered as one of the most promising thermochemical technologies and experimental results are presented for a fluidized bed [1] and downdraft [2] gasifiers.

The complete technology from solid fuel to final application involves several unit operations including grinding of fuel, drying, gasification, gas cooling, gas cleaning and final utilization of the gas. It was previously demonstrated by Vlaswinkel [3] for gasification of coal and recently by Ptasinski et al. [4] for gasification of organic sludge that the gasifier is one of the least-efficient unit operations in the whole gasification technology. Therefore, an analysis of the gasifier alone can substantially contribute to the efficiency improvement of the whole gasification technology.

The main purpose of this paper is to compare different types of biofuels for their gasification efficiency and benchmark this against gasification of coal. A wide range of biomass sources, such as traditional agricultural crops, dedicated energy crops, residues from agriculture and foresting as well as organic wastes can be gasified. This is generally regarded as a real advantage, because it means that the best available and usually economically most

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Nomenclature

C_S	calorific value of sulphur (kJ/kg)
ΔH	enthalpy of reaction (kJ/mol)
LHV	lower heating value (kJ/kg)
LHV_{org}	LHV of organic fraction in biomass per kilo gram of organic fraction (kJ/kg org.fr.)
LHV_{org}	LHV of organic fraction in biomass per kilo gram of wet biomass (kJ/kg biomass)
T	temperature (°C)
z	mass fraction (dimensionless)
β	ratio of the chemical exergy to the LHV of dry organic substances (dimensionless)
ε_{ch}	chemical exergy (kJ/kg)
ε_{ph}	physical exergy (kJ/kg)
Ψ	exergy efficiency (dimensionless)

attractive feedstock can be selected. However, these biomass types differ in chemical composition, heating value, ash and moisture content. The question is whether all the biomass types can be converted with comparable efficiencies. Biomass is not a well-defined and often inhomogeneous feedstock, whose composition may vary depending on origin, physical location, age, season and other factors. The composition of the organic matter in biomass does not vary much, e.g., wood typically contains cellulose:hemicellulose:lignin in 2:1:1 ratio. However, variations in moisture content and ash content are especially large. In this study various fuels, comprising wood, grass and vegetable oils and including wet biomass sources such as manure and sludge are studied. Coal was also chosen as a reference point for fossil fuel gasification.

The change in composition from biomass to coal can be illustrated using a diagram developed by Van Krevelen [5]. Fig. 1 shows the change in atomic ratios H/C and O/C from biomass to peat, lignite, coal and anthracite. As it can be observed from this figure, biomass, e.g., wood, contains more oxygen with respect to carbon than in coal. This influences the gasification process, as gasification is a partial oxidation.

This study focuses on the comparison of different fuels for their gasification efficiency. To this end the idealized gasifier model is used with the following assumptions: (i) the chemical equilibrium between gasifier products is reached, (ii) the ashes are not considered and (iii) heat losses in the gasifier and dryer are neglected.

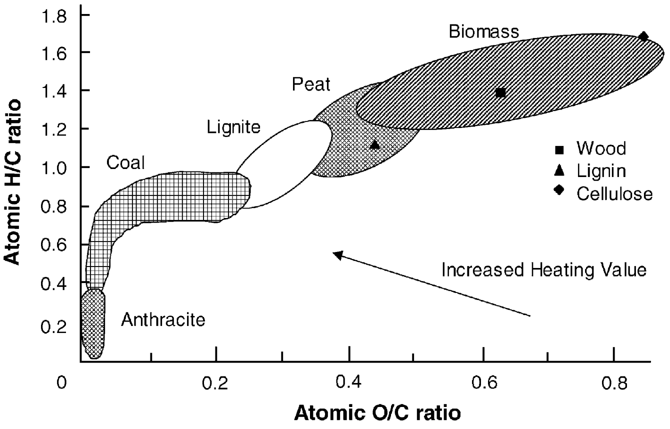

Fig. 1. Van Krevelen diagram.

This paper starts with an evaluation of exergetic efficiencies. Focusing thereafter on biomass gasification, the question is under which gasification conditions the gasification efficiencies should be evaluated. It was shown by Desrosiers [6] and Double and Bridgwater [7] that energy-based efficiencies reach a sharp maximum at the so-called carbon-boundary point where exactly the right amount of oxygen is added to the gasifier in order to achieve complete gasification. Prins et al. [8] have shown that the carbon-boundary point is also the optimum for exergy-based efficiencies. The locations of the carbon-boundary points with corresponding carbon-boundary temperatures and gas phase compositions are calculated for a variety of biofuels. The results are presented for gasification efficiencies of these fuels, based on energy and exergy. Finally, the effect of improving gasification of wet biomass by prior drying using the enthalpy of the product gas is evaluated.

2. Evaluation of exergy efficiency

The gasification process considered in this paper is schematically presented in Fig. 2. Biomass enters the gasifier at environmental temperature T_0, air at the same temperature is used as a gasifying medium. The gaseous products leave the gasifier at the reactor temperature T_R. It is assumed that the gasifier operates as adiabatic and pseudo-homogeneous reactor at atmospheric pressure. Gasification entails partial oxidation of the feedstock, so chemical energy of biomass is converted into chemical and thermal energy of synthesis gas.

The energetic efficiency of a gasification process, generally known as the cold-gas efficiency, can be

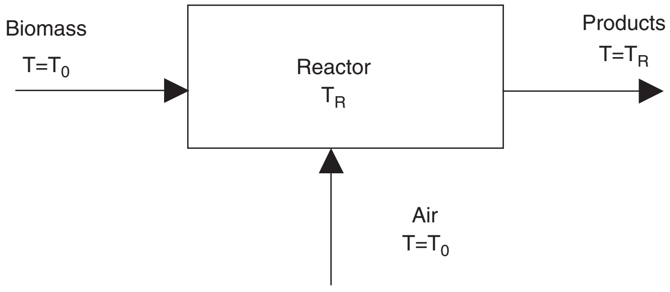

Fig. 2. Schematic diagram of the gasifier.

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determined as

$$\Psi_1 = \frac{\mathrm{LHV_{gas}}}{\mathrm{LHV_{biomass}}}, \tag{1}$$

where $\mathrm{LHV_{gas}}$ and $\mathrm{LHV_{biomass}}$ are the net heats of combustion (lower heating values) of gas and biomass, respectively. The lower heating values for the organic fraction of the biomass were calculated from the higher heating values, present in the Phyllis database, maintained by the Energy Research Centre of the Netherlands (ECN) [9].

The exergetic efficiency may be defined as the ratio between chemical exergy of product gas and biomass feed

$$\Psi_2 = \frac{\varepsilon_{\mathrm{ch,\,gas}}}{\varepsilon_{\mathrm{ch,\,biomass}}}. \tag{2}$$

Both definitions have a drawback: they disregard the sensible heat contained in the product gases. Synthesis gas at elevated temperature is preferred over environmental temperature and therefore the calculated efficiency should be higher. However, if the sensible heat were to be added to the energetic efficiency, this efficiency would always be 100% because the gasifier operates adiabatically. This problem can be overcome by using exergetic efficiency based on chemical as well as physical exergy

$$\Psi_3 = \frac{\varepsilon_{\mathrm{ch,\,gas}} + \varepsilon_{\mathrm{ph,\,gas}}}{\varepsilon_{\mathrm{ch,\,biomass}}}. \tag{3}$$

Exergy analysis is performed according to the method proposed by Szargut et al. [10]. Chemical exergy of the biomass is calculated from the correlations for technical fuels using the LHV, and mass fractions of organic material, sulphur, water and ash in the biomass

$$\begin{aligned}\varepsilon_{\mathrm{ch,\,total}} &= z_{\mathrm{org}}(\beta\,\mathrm{LHV_{org}}) + z_{\mathrm{S}}(\varepsilon_{\mathrm{ch,\,S}} - C_{\mathrm{S}}) \\ &\quad + z_{\mathrm{water}}\,\varepsilon_{\mathrm{ch,\,water}} + z_{\mathrm{ash}}\varepsilon_{\mathrm{ch,\,ash}}.\end{aligned} \tag{4}$$

The factor β is the ratio of the chemical exergy to the LHV of the organic fraction of biomass. This factor is calculated from statistical correlations developed by Szargut and Styrylska [11]. The following correlation is used:

- for solid biofuels:

$$\beta = \frac{1.044 + 0.0160\,\mathrm{H/C} - 0.3493\,\mathrm{O/C}\lfloor 1 + 0.0531\,\mathrm{H/C}\rfloor + 0.0493\,\mathrm{N/C}}{1 - 0.4124\,\mathrm{O/C}}, \tag{5}$$

- for liquid vegetable oils:

$$\beta = 1.0374 + 0.0159\frac{\mathrm{H}}{\mathrm{C}} + 0.0567\frac{\mathrm{O}}{\mathrm{C}}, \tag{6}$$

- for coal:

$$\begin{aligned}\beta &= 1.0437 + 0.1869\frac{z_{H_2}}{z_C} + 0.0617\frac{z_{O_2}}{z_C} \\ &\quad + 0.0428\cdot\frac{z_{N_2}}{z_C},\end{aligned} \tag{7}$$

where H/C, O/C, and N/C represent atomic ratios in the fuel.

3. Gasification of various biofuels at the carbon-boundary point

Table 1 shows proximate and ultimate analyses for the fossil fuel and the biofuels that were considered. The presented data were obtained from the Phyllis database [9]. Since biomass and (to a lesser degree) coal contain mainly carbon, hydrogen and oxygen, the compositions of the fuels can be indicated in a molar ternary C–H–O diagram as shown in Fig. 3. The equilibrium composition at any point in the diagram can be computed at a given temperature and pressure by minimization of the Gibbs free energy of the system. In this work, only thermodynamically stable components at temperatures above 600 °C were considered, i.e. CO, CO_2, H_2, H_2O, CH_4, C(s), N_2 and H_2S. The main chemical reactions taking

Table 1
Proximate and ultimate analysis of various fuels

Fuel	Proximate analysis (wt%)			Ultimate analysis (wt% of organic fraction)				
	Moisture	Ash	Organic fraction	C	H	O	N	S
Coal	11.5	8.50	80.0	78.2	4.93	13.3	1.45	1.69
Vegetable oils	0.0	0.0	100.0	75.4	11.7	12.9	0.0	0.0
Straw	12.7	6.37	80.9	48.9	5.97	43.9	0.82	0.15
Treated wood	14.6	4.44	81.0	51.5	6.03	41.3	1.22	0.09
Untreated wood	19.8	1.84	78.4	50.8	6.06	42.7	0.36	0.07
Grass/plants	24.2	5.46	70.3	49.7	6.00	42.7	1.32	0.18
Sludge	32.5	25.72	41.8	50.2	7.09	34.9	5.63	1.77
Manure	43.6	17.20	39.2	50.2	6.50	34.6	5.19	0.85

Note: Ultimate analysis does not add up to exactly 100% due to presence of trace elements (e.g., chlorine).

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place in the gasifier are:

$$C + CO_2 = 2CO,\ \Delta H = 172.6\,\text{kJ/mol} \tag{8}$$

$$C + H_2O = CO + H_2,\ \Delta H = 131.4\,\text{kJ/mol} \tag{9}$$

$$C + 2H_2 = CH_4,\ \Delta H = -74.9\,\text{kJ/mol} \tag{10}$$

$$C + O_2 = CO_2,\ \Delta H = -393.5\,\text{kJ/mol} \tag{11}$$

In Fig. 3, the indicated isotherms are the solid carbon-boundary lines (at a temperature of 600 and 832 °C). Above the carbon-boundary line, solid carbon exists in heterogeneous equilibrium with gaseous components; below this line, only gaseous components are present in homogeneous equilibrium. Gasification of fuel implies adding so much oxygen until the solid carbon boundary is reached. This is indicated for coal by the arrow in Fig. 3, which points in the direction of oxygen; when point A is reached, all carbon has been gasified. The carbon-boundary point is the optimum point of the gasifier operation because exactly enough oxygen is added to achieve complete gasification. If more oxygen is added than required, the produced gas loses its heating value, until eventually the line from CO_2 to H_2O is crossed and complete combustion has taken place.

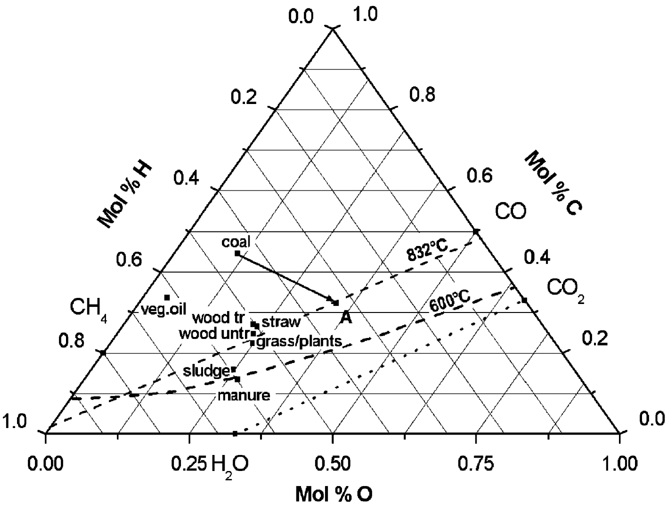

Fig. 3. Location of biofuels in ternary C–H–O diagram.

For the various biofuels considered, the optimum point of gasifier operation was calculated by the software program Aspen Plus assuming that chemical equilibrium is attained. Table 2 shows the corresponding air/biomass ratios and carbon-boundary temperatures for the optimum gasification points for the fuels. From these data, it can be observed that the solid biofuels require much less addition of air than coal or vegetable oil. The reason for this is that they already contain a large amount of oxygen in their organic matter. Since relatively little oxygen has to be added to the biomass, less oxidation reactions take place and the corresponding carbon-boundary temperatures for gasification of biomass are lower than for coal and vegetable oil.

The compositions of sludge and manure lie already very close to the carbon-boundary isotherm of 600 °C. Because sludge and manure are very moist, they already contain a large amount of oxygen, so that oxygen addition is limited, and carbon-boundary temperatures were found to be below 600 °C. However, at such low temperatures, the gasification reactions become very slow and it would be difficult to attain chemical equilibrium in practice. A gasification temperature of at least 600 °C was considered as being required in order to reach equilibrium within a reasonable reaction time. This criterion dictated the amount of air for gasification of sludge and manure, which means that these fuels are over-oxidized in order to evaporate all the moisture present.

Table 2 shows the composition of the products formed by the gasification process. As a result of the higher gasification temperature for coal and vegetable oil, more carbon monoxide and hydrogen are formed, while gasification of the other biofuels takes place at lower temperatures and the gas contains more carbon dioxide and methane. From the table, it is apparent that very moist gas streams result from gasification of sludge and manure.

4. Results of exergy analysis

Table 3 shows lower heating values, the ratio β, and total chemical exergy of the various fuels. It is remarkable that

Table 2
Optimum gasification operating points and gas compositions for various fuels

Fuel type	T (°C)	Air flow (kg/kg biomass)	Product gas composition (mole fraction)						
			H_2O	N_2	H_2	CO	CO_2	CH_4	H_2S
Coal	832	2.836	0.005	0.500	0.158	0.324	0.009	0.001	0.003
Vegetable oils	875	3.837	0.003	0.467	0.251	0.275	0.003	0.001	0.000
Straw	659	1.401	0.063	0.384	0.225	0.205	0.113	0.010	0.000
Treated wood	655	1.628	0.062	0.409	0.213	0.194	0.112	0.010	0.000
Untreated wood	642	1.452	0.076	0.380	0.227	0.177	0.126	0.013	0.000
Grass/plants	621	1.240	0.097	0.363	0.232	0.146	0.145	0.018	0.000
Sludge[a]	600	1.237	0.186	0.412	0.192	0.056	0.147	0.004	0.003
Manure[a]	600	1.247	0.246	0.395	0.171	0.038	0.147	0.002	0.001

[a] For this feedstock, the carbon-boundary temperature is below 600 °C. A minimum gasification temperature of 600 °C was used.

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Table 3
Lower heating value and chemical exergy of various fuels

Fuel	LHV$_{org}$ (kJ/kg organic)	LHV$_{org}$ (kJ/kg biomass)	β (−)	ε_{ch} (kJ/kg biomass)
Coal	31047	24839	1.067	26638
Vegetable oils	37558	37558	1.074	40338
Straw	18064	14619	1.128	16506
Treated wood	18886	15290	1.119	17129
Untreated wood	18904	14812	1.122	16634
Grass/plants	18624	13101	1.125	14760
Sludge	19617	8197	1.118	9249
Manure	19148	7506	1.116	8427

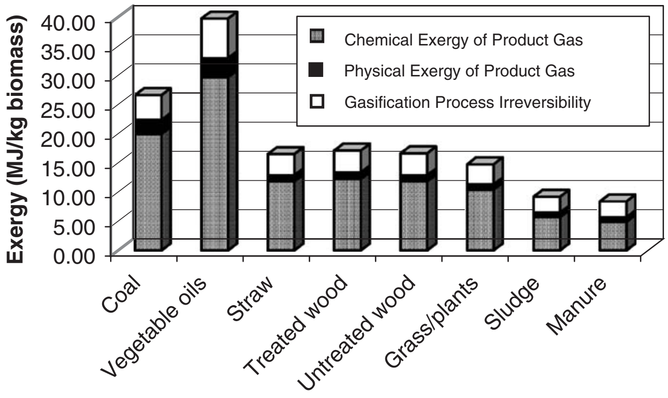

Fig. 4. Exergy distribution for gasification of various fuels.

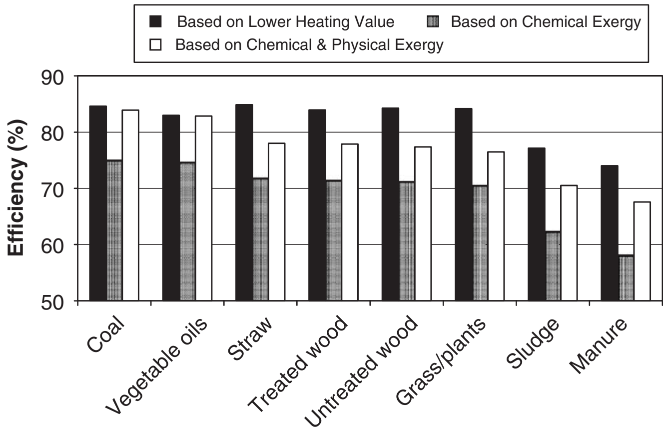

Fig. 5. Comparison of gasification efficiency for various fuels.

for biomass containing a lot of oxygen, the values of β are higher (above 1.10) than for coal and vegetable oils (around 1.07) with much lower oxygen content. This may be explained by the fact that polymers such as cellulose and hemi-cellulose are highly ordered structures, and work can be delivered if these are decomposed.

Fig. 4 shows the exergy distribution for gasification of various biofuels. The exergy contained in the biomass is converted by gasification into chemical exergy of the product gas, physical exergy of the product gas, and part of the exergy of biomass is lost due to process irreversibilities. It can be seen that the largest irreversibility occurs for vegetable oil and coal gasification, which is related to the large amount of oxygen that needs to be added to vegetable oil and coal gasifiers. However, rather than the absolute irreversibility, it is more meaningful to focus on relative irreversibility, i.e., to compare the biofuels due to exergetic efficiencies. The absolute irreversibility indicates the amount of feedstock exergy being lost in the gasification process, whereas the exergetic efficiency shows the ratio between the exergy of useful gasification products and the feedstock exergy. This way different fuels can be compared based on the degree of utilization of their chemical exergy.

Fig. 5 shows the energetic and exergetic efficiencies for the gasification of different biofuels. The energetic efficiencies of vegetable oil, straw, treated wood, untreated wood and grass are very comparable with coal, whereas efficiencies for sludge and manure are considerably lower. This was to be expected, because gasification at the optimum operating point was not possible for these streams. In sludge and manure gasification, oxygen is added mainly to generate heat and evaporate moisture present in the fuel. If the exothermic oxidation reactions could drive endothermic gasification reactions, rather than endothermic evaporation of water, the gasifier would work much more efficiently.

Fig. 5 shows that the efficiency based on chemical exergy is higher for coal and vegetable oil than for the other biomass streams, i.e., around 75% vs. 70–72%. This may be explained because large molecules are broken up into smaller ones. For some of the small molecules contained in the product gas, the chemical exergy is less than their lower heating value e.g., 97.6% for hydrogen and 97.2% for carbon monoxide. On the contrary, the chemical exergy for coal and biomass is higher than their heating value. Therefore, the difference in chemical exergy between feed and product is larger than the difference in lower value. This difference is smaller for coal and vegetable oils, because their chemical exergy is only 107% of their lower heating value, while for solid biomass this is 111–113%. Also, exergetic efficiencies of gasification are lower than energetic efficiencies.

Gasification efficiencies based on chemical and physical exergy are also shown in Fig. 5. The same trends are observed as before: gasification of vegetable oil and coal is

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better than biomass such as wood, straw or grass, while gasification of manure and sludge is much less efficient. Because coal and vegetable oil are gasified at higher temperatures, their gasification efficiencies are improved relatively much by inclusion of the physical exergy. Drier biomass such as treated wood or straw may be slightly preferred over fresh biomass such as untreated wood and grass.

Finally, it is very interesting to note that gasification of vegetable oils, which are moisture and ash-free, is comparable to gasification of coal, despite containing moisture and ash. It can be noted that chemical exergy of vegetable oils and coal is higher than that of remaining fuels (see Table 3).

5. Improving gasification of wet biomass

It was concluded that gasification of sludge and manure is not very efficient because these contain a substantial amount of water, so that operation at the optimal gasification conditions is not possible. This is caused because extra heat must be generated at the gasification temperature ($>600\,^{\circ}C$) in order to evaporate the water at a much lower temperature, leading to exergetic losses. In order to improve the gasification efficiency, moisture should be removed prior to the gasifier. It is possible to use the enthalpy of the gasifier product gas for drying the biomass (in the absence of alternative, external sources of heat). We have assumed that water is evaporated in the dryer at a temperature of 100 °C by exchanging heat with the product stream, as shown in Fig. 6. Modelling of drier and gasifier was restricted to sludge, as this was one of the two streams containing relatively much moisture. In this new situation, the temperature of the products is lower than the temperature of the gasifier, due to the fact that heat is removed for the vapourization of the water.

Calculations were performed using Aspen Plus, and the sludge was dried from its initial moisture content of 32.5% to water percentages varying from 30% to 11%. This decreases the final temperature of the product gas from 600 °C to temperatures between 514 and 297 °C. It was found that gasification of dried sludge containing 19 wt% moisture corresponded to a carbon-boundary temperature of 600 °C. Therefore, by drying the sludge from 32.5 wt% moisture to 19 wt% using product gas, it was possible to enable gasification at the point where not more oxygen is added than required for gasification of the carbon. This is demonstrated in the ternary C–H–O diagram in Fig. 7. By gasification of the original wet biomass at 600 °C, point B is reached which is below the carbon-boundary isotherm. If water is removed from the sludge by prior drying, gasification at the carbon boundary becomes possible, indicated by moving from the point indicated with 19% moisture to point A on the carbon-boundary line.

Fig. 8 shows the effect of drying the biomass on the overall efficiency of the gasification system, including the dryer. It is obvious that the efficiency improvement by drying is less for the efficiency based on the total exergy compared to the efficiencies based on the lower heating value or the chemical exergy. The reason is that this definition takes the temperature decrease of the products into account. As the moisture content decreases, higher amounts of water have to be evaporated in the dryer. Because the heat needed to vapourize this water is removed from the hot gases leaving the gasifier, the temperature of

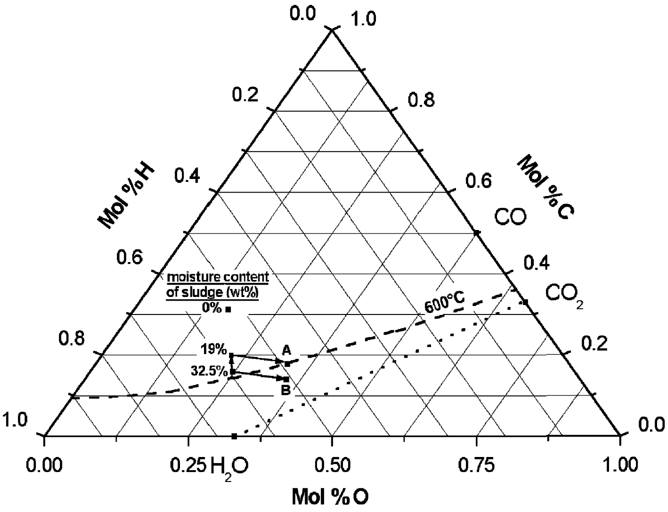

Fig. 7. Drying and gasification of sludge illustrated in ternary C–H–O diagram.

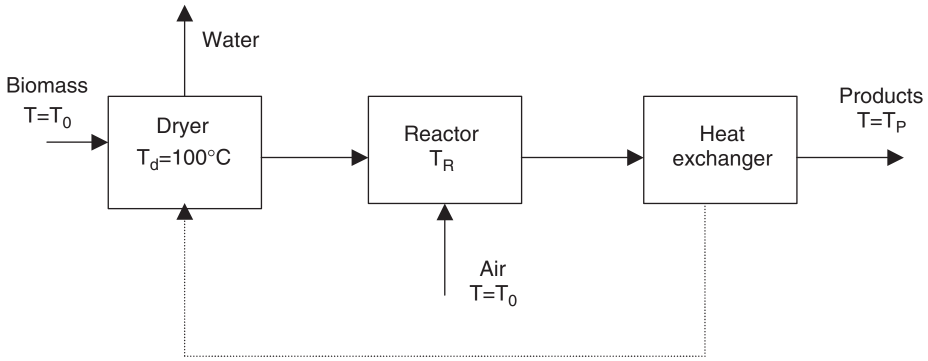

Fig. 6. Process scheme for drying of biomass prior to the gasification process.

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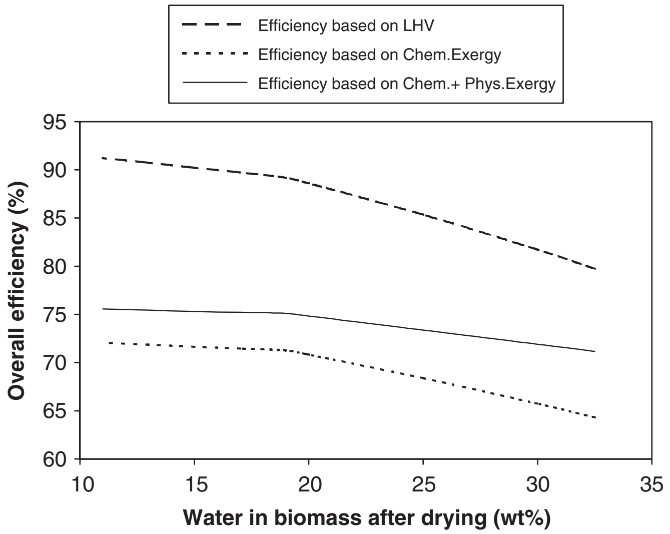

Fig. 8. Improvement of gasification efficiency by drying using enthalpy of product gas.

the products decrease, thus lowering the value of the physical exergy.

As expected, decreasing the moisture content of the biomass entering the gasifier increases the efficiencies of the process. The largest benefit is obtained when the sludge is dried from 32.5% to 19% moisture. Further drying hardly increases the chemical exergy of the product gas, but the physical exergy of the product gas is slightly increased because carbon-boundary temperatures higher than 600 °C can be achieved. Therefore, at increased drying levels (that is for water content in the biomass leaving the dryer lower than 19%), the efficiency based on chemical and physical exergy has the same slope as the efficiency based only on chemical exergy. Moreover, at this range the efficiency based on the total exergy (chemical and physical exergy) is only slightly higher than the efficiency based only on the chemical exergy. The reason is that the chemical exergy of the biomass is the main constituent of the total exergy and the contribution of physical exergy is much smaller.

6. Conclusions

In order to substitute fossil fuels by renewable fuels, solid biofuels (straw, untreated wood, treated wood, grass/plants) or liquid biofuels (vegetable oil) could replace coal as a gasification feedstock. The optimum gasification efficiencies of these fuels based on lower heating values are comparable, i.e., around 84%. However, if the efficiencies are based on chemical exergy, the solid biofuels with high oxygen content are regarded as high-quality fuels, for which a penalty is paid when decomposing them into small gaseous components. Also, the gas produced from solid biomass gasification has a lower temperature so that it contains less physical exergy. As a result of these two factors, gasification based on chemical and physical exergy shows higher efficiencies for coal than for solid biomass, i.e., almost 84% vs. 76–78%. It is interesting to note that gasification of vegetable oils is similar to the gasification of coal, and both these fuels can be considered as high-quality fuels.

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