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Synthesis, Modeling and Exergy Analysis of Atmospheric Air Blown Biomass Gasification for Fischer-Tropsch Process*

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Abstract

The main objective of the present work is to investigate the combination of an atmospheric and a pressurised air blown biomass gasifier towards Fischer-Tropsch (FT) synthesis of second generation biofuels and co-production of power. The complete process was simulated in AspenPlus^{TM} software including sub-models for the gasification, gas cleaning and conditioning as well as for the FT reactor. A sensitivity analysis on exergetic performance was performed mainly examining the recycling of FT-tail gas. For the base case of 80% FT CO conversion without any recirculation, the exergetic efficiency was found to be 34.3% with atmospheric gasification and 30.64% for pressurised gasification. Recirculation of the non-reacted CO, H_2 as well as light gas products of the FT-reactor back to the atmospheric gasifier can increase the overall efficiency up to 48.1 %.

Keywords: *Biomass gasification, Fischer–Tropsch, exergy, AspenPlus^{TM}.*

1. Introduction

Currently transportation sector consumes more than 20% of the primary energy worldwide and is estimated to reach up to 30% by 2050 (Hamelinck et al., 2006). The increase of oil and natural gas prices as well as the uncertainty on their availability drives efforts to substitute fossil transportation fuels. Amongst the renewable sources, only biomass is directly available to contribute towards this aim. According to an EU Directive 2003/30/EC (2003), the target for the share of liquid biofuel for 2020 is 15%. Some 5% could be achieved by first generation biofuels (biodiesel and bioethanol), while the rest should be possible via second generation biofuels.

Fischer-Tropsch (FT) synthesis is a known technique for the production of liquid transportation fuel with significant industrial applications, using mainly coal and natural gas, (Espinosa et al., 1999; Steynberg et al., 1999; Tijmensen et al., 2002) and is presented to be one of the most promising techniques for the production of second generation biofuels. FT products are free of S and N compounds, have a very low aromatic hydrocarbons content and high cetane numbers, resulting in reduced emissions when used in internal combustion (Tijmensen et al., 2002; Dry, 2001). The FT-products are compatible with the existing fuel infrastructure, in contrast with competitive biofuels (Hamelinck et al., 2004). The cost of FT-products is estimated to be in the range of 15 € GJ^{-1}, approximately 40% more expensive than bioethanol, but could be reduced to the range of 10 € GJ^{-1} with further research and increase of the capacity of the so called biorefineries (Hamelinck et al., 2004).

The main objective of the present work is to investigate the combination of an atmospheric air blown biomass gasifier with Fischer-Tropsch (FT) synthesis for the co-production of liquid biofuel and power. This option has not been investigated thoroughly but offers the possibility for increased efficiencies by reusing FT tail gas in the low pressure gasifier.

2. System Configuration

A significant part of carbon species remains in the tail gas of the FT process, reducing significantly the liquid productivity (Tijmensen et al., 2002;Hamelinck et al., 2004; Boerrigter et al., 2002; Prins et al., 2005). In the proposed configuration, shown in Figure 1, emphasis that should be paid is in the optional employment of membranes to separate and direct part of the FT-tail gas back into the gasifier. In more detail, the proposed system flowchart consists of an atmospheric air blown biomass gasifier, a product gas cleaning and conditioning system, including a three stage compressor with intercooler and a WGS (Water Gas Shift) reactor, and a fixed bed FT reactor. Heat from the cooling of the product gas as well as from the FT-synthesis is recuperated and used for the production of steam which produces power with a Rankine cycle. Table 1 summarizes the operating parameters of the major unit operations comprising the Aspenplus^{TM} flow sheet modelling used for the evaluation of the system's efficiency.

Table 1. Operating Parameters for Different Components.

Gasifier	$T = 1073$ K,	$\lambda = 0.33$ (air ratio)
Air Blower	$\eta_{isentr} = 0.75$	$\eta_{mech} = 0.99$
Compressors	$\eta_{isentr} = 0.80$	$\eta_{mech} = 0.99$
WGS Reactor	$T = 523$ K	$P = 26$ bar
FT Reactor	$T = 523$ K	$P = 25$ bar
	$\Delta P = 5$ bar	
	CO-conv. = 0.4/ 0.6/ 0.8	
Steam Turbine	$\eta_{isentr} = 0.85$	$\eta_{mech} = 0.99$
Heat Exchangers	5% loss	$\Delta T = 25$ K

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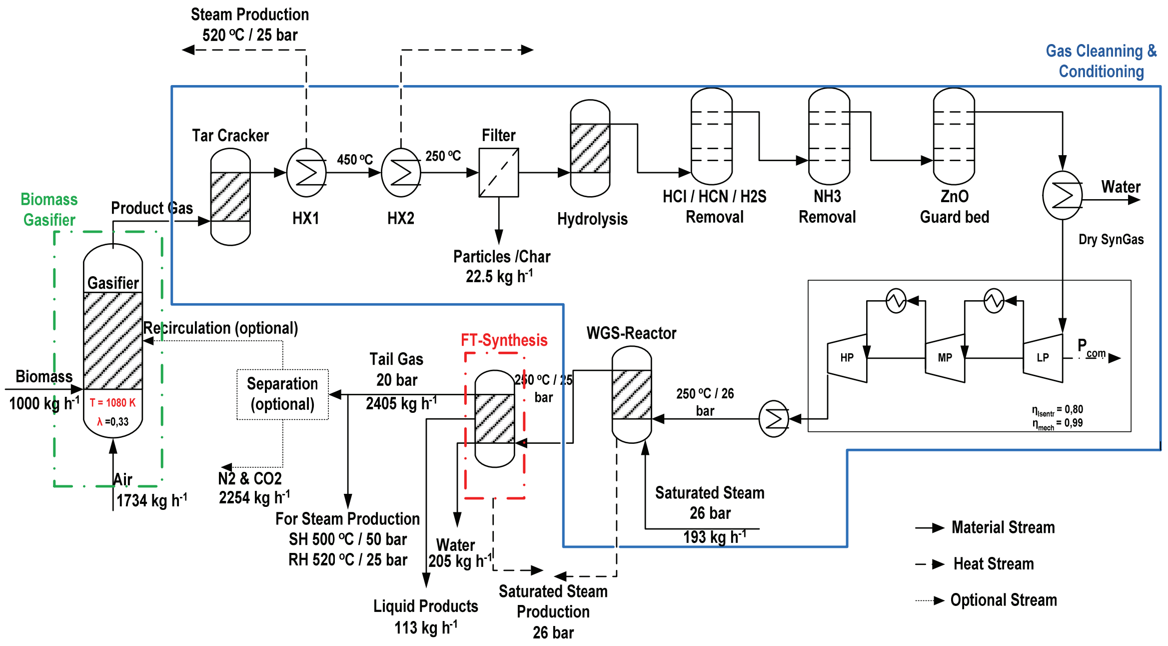

Figure 1. FT Production Process Flowchart (with base case flows).

The system was further analysed exergetically by following the methodology described in paragraph 3.1. Separate exergetic efficiencies were defined for the major unit operations i.e. gasification, gas cleaning and conditioning and the FT reactor and are presented in 3.2, 3.3, and 3.4 respectively.

3. Exergy Analysis

3.1 Methodology

The total exergy of a material stream is expressed as the sum of two components:

$$E = N\left(\varepsilon_{ph} + \varepsilon_{ch}\right) \tag{1}$$

where molar physical exergy is:

$$\varepsilon_{ph} = (h - h_o) - T_o(s - s_o) \tag{2}$$

and molar chemical exergy for an ideal mixture/ solution:

$$\varepsilon_{ch,gas} = \sum_i x_i \varepsilon_{oi} + \mathrm{R}T_o \sum_i x_i \ln x_i \tag{3}$$

Potential, kinetic exergies etc were neglected.

Mole flows and molar fractions, the enthalpy and entropy of material streams were taken from the AspenPlus^{TM} flow sheet results; for each material stream an identical one was created and brought to standard environmental conditions (T_o=298.15 K, p_o=1.013 bar) so as to evaluate its reference molar enthalpy and entropy (h_o, s_o). Values of ε_{oi} were taken from Kotas (1995), as the chemical exergy of biomass was calculated using the statistical correlation β for wood, proposed by Szargut et al. (1988) for solid fuels (Woody biomass data are given in Table 2).

$$\varepsilon_{ch,biom} = (LHV_{fuel} + wh_{fg})\beta \tag{4}$$

Exergy of power equals the rate of power and exergy of a heat stream Q was evaluated using Carnot factor:

$$E_T^Q = Q(1 - T_o / T) \tag{5}$$

where T is the temperature at which Q is available.

The total exergy balance of a steady state process has the following form:

$$\sum_{IN} E_j + \sum_{IN} E_{T_{IN}}^Q = \sum_{OUT} E_k + \sum_{OUT} E_{T_{OUT}}^Q + \sum_{OUT} E^W + I \tag{6}$$

where the irreversibility I represents the loss of quality of materials and energy due to dissipation.

Table 2. Biomass Fuel Data.

Proximate analysis (% w/w dry)		Ultimate analysis (% w/w dry)	
Volatiles	83.20	C	50.10
Fixed Carbon	16.17	H	6.12
Moisture	10.0	O	43.06
Heating Values d.b.		N	0.06
HHV (kJ/kg)	18900	S	0.02
LHV (kJ/ kg)	15567	Cl	0.01

3.2 Biomass Gasification

The gasification control volume is shown in Figure 2.

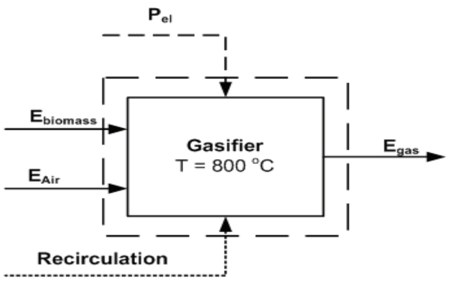

Figure 2. Gasification Control Volume.

Gasification process was modelled in AspenPlus^{TM} in a simple manner to be accurate in providing product gas main composition. The main assumptions for the species that are not predicted by thermodynamics were: a) unreacted char (modelled as graphite $C_{(s)}$) was 5% w/w of the biomass carbon input, and b) CH_4 concentration in the product gas was assumed 4% vol. in N_2-free basis for near atmospheric gasification and 14% N_2-free basis for the pressurised air gasification (Kakaras et al. 2003; Stahl et al. 1999; Stahl et al. 2004). Tars have been modelled as naphthalene ($C_{10}H_8$) with a load of 5 g Nm^{-3}. The remaining major gas constituents were assumed at thermodynamic equilibrium (H_2, CO, CO_2, and H_2O).

The operating temperature of the gasifier in both cases was chosen T_{gas}= 800°C in order to avoid bed agglomeration problems (Fryda et al., 2008). The operating pressure for the atmospheric air gasifier was chosen p_{gas} = 1.4 bar, enough to overcome subsequent pressure drop in the gasifier, the gas cleaning system and heat exchangers. The operating pressure for the pressurised gasifier was chosen p_{gas} = 27 bar, and a pressure drop of 2 bar in the gas cleaning and conditioning system as well as in the heat exchangers, was assumed. Exergetic efficiency takes into account the entropy increase due to conversion of a solid to a gaseous fuel (Prins et al. 2003) based on the definition of the degree of perfection for a process by Szargut (1988), the exergetic efficiency for the gasification is expressed:

$$\eta_{ex,gas} = \frac{E_{Gas}}{E_{Biomass} + E_{Air} + E_{Recirc} + E^{W}} \tag{7}$$

For the case where there is no recirculation of tail gas (E_{Recirc} = 0), the exergetic efficiency of atmospheric air gasification is evaluated 76.79%.

3.3 Gas cleaning and Conditioning

Product gas cleaning for FT synthesis application is significantly more demanding compared with conventional application (i.e. Internal Combustion Engines, GT). For commercial scale application, the exact limits for gas cleaning are determined based on economic considerations: investment cost of gas cleaning system versus cost from loss of production and replacement of the catalyst. Different concentrations limits have been proposed in the literature (Turk et al. 2001; Tijmensen, 2002; Hamelinck, 2004; Boerrigter, 2002).

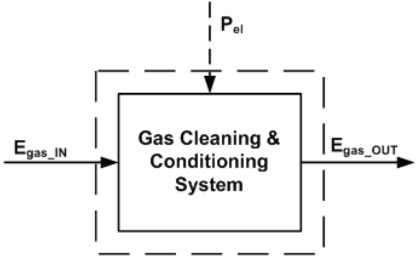

Figure 3. Control Volume for Gas cleaning and Conditioning System.

The control volume for the gas cleaning and conditioning system (GCCS) is shown in Figure 3. A warm gas cleaning, illustrated in Figure 1, is proposed, consisting of a catalytic tar cracker reactor, a ceramic filter for particles, a hydrolysis reactor and four sorbent-bed reactors for the removal of NH_3, HCl and H_2S. In the present work extensive scavenging of the contaminants is assumed.

The cleaned product gas is then cooled down, compressed up to 25 bar, using a three stage compressor with intercoolers and shifted in a catalytic WGS reactor, so as to arrange the H_2/CO molar ratio around 2.1.

The exergy efficiency of the gas cleaning and conditioning is defined as:

$$\eta_{ex,gccs} = \frac{E_{Gas,OUT}}{E_{Gas,IN} + E^{W}} \tag{8}$$

Raw product gas is defined as the incoming and WGS reactor outlet as the outgoing streams. For the base case (no tail gas recirculation), the exergetic efficiency is evaluated at 87.22%.

3.4 Fischer-Tropsch Reactor

The FT synthesis is a carbon chain building process, where CH_2 groups are attached in a carbon chain. The resulting overall reaction can be presented as:

$$n\mathrm{CO} + 2n\mathrm{H}_2 \rightarrow [-\mathrm{CH}_2\text{-}] + n\mathrm{H}_2O \quad \Delta H^{o}_{\mathrm{FT}} = -165kJ/mol$$

In the present work only the above mentioned reaction is taken into consideration, although other reactions also occur (Rauch, 2003).

A cobalt catalyst fixed bed FT reactor was modelled. In order to evaluate the FT HC product distribution, a FORTRAN block was created and integrated to Aspenplus^{TM} modelling, based on Anderson-Schulz-Flory (ASF) distribution as described in Perry (1999):

$$\log\left(\frac{W_n}{n}\right) = \mathrm{n}\ \log\alpha + \log\left[\frac{(1-\alpha)^2}{\alpha}\right] \tag{9}$$

For the calculation of the probability α, the Eq. (10) was used (Prins et al., 2005; Vessia et al., 2005):

$$\alpha = \frac{\mathrm{k_{HC1}} \cdot P_{\mathrm{CO}}}{\mathrm{k_{HC1}} \cdot P_{\mathrm{CO}} + \mathrm{k_{HC5}} \cdot P_{\mathrm{H_2}} + \mathrm{k_{HC6}}} \tag{10}$$

where, P_{CO} and P_{H2} are the partial pressures of CO and H_2 in bar, k_{HC1} is the rate constant for the adsorption of carbon monoxide on an active site and equals with $1.22.10^{-5}$ mol(g s bar)$^{-1}$, k_{HC5} is the rate constant for desorption of paraffin by hydrogenation of active site, and equals with $1.05.10^{-6}$ mol(g s bar)$^{-1}$ and k_{HC6} is the rate constant for desorption of olefins from active site, and equals with $2.36.10^{-6}$ mol(g s)$^{-1}$. These values were taken from Vessia et al. (2005). The FT-product distribution defined by the Anderson-Schulz-Flory is presented in Figure 4.

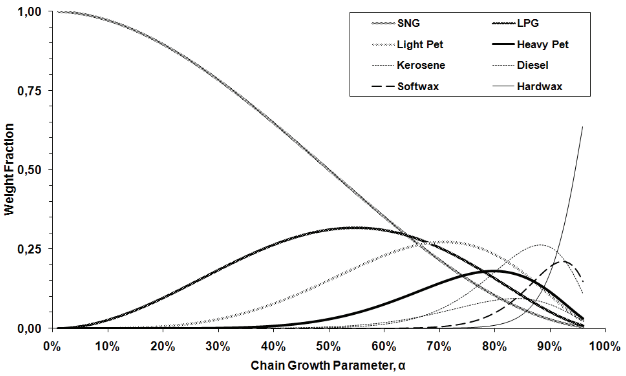

Figure 4. FT Product Distribution According to ASF Distribution.

Figure 5 illustrates the FT-product distribution for the base case of atmospheric gasification and no re-circulation.

FT synthesis process heat surplus is used to produce saturated steam at 26 bar part of which is fed to the WGS reactor while the rest is contributing to a power producing Rankine cycle.

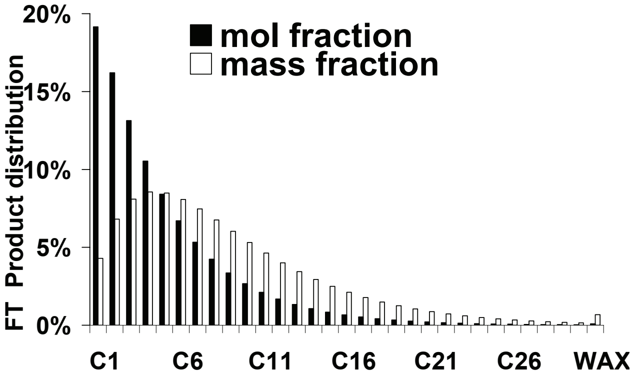

Figure 5. FT Product Distribution for the Base Case (atmospheric air gasification with FT CO-conversion 80%, @ P_{tot} = 25 bar).

The FT products are cooled in order to separate the liquid hydrocarbons from lighter gases (tail gas). The FT reactor control volume is presented in Figure 6, as its exergy efficiency is defined as:

$$\eta_{ex,FT} = \frac{E_{biof} + E^{Q}_{523K}}{E_{Gas,IN}} \qquad (11)$$

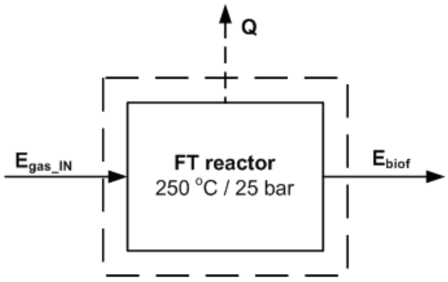

Figure 6. Control Volume FT Reactor.

E_{biof} corresponds to the liquid FT products. For the base case (no recirculation of tail gas) the exergetic efficiency of the FT reactor is 43.23%, 32.94% and 18.53% assuming a CO-conversion of 80%, 60% and 40%, respectively.

4. Results and Discussion

4.1 Base Case Results

The energy and exergy efficiencies and the conversion of biomass carbon to liquid biofuels are presented in Table 3 for different CO-conversion rates in the FT reactor. The biofuel production increases with the conversion rate while the power production is mainly depending on the total exergy content of the tail gas is reduced.

Comparing the different types of gasifiers used, atmospheric gasification is more efficient in the production of liquid biofuels. The main reason is the significant higher concentration of CH_4 in the product gas of pressurized gasification, which is inert in the FT-synthesis. As a consequence of the high CH_4 concentration of the product gas, the net power production is significantly increased in pressurized gasification mode. Almost similar work is required to pressurize the air for pressurized gasification compared with the work required to pressurize the atmospheric product gas. Therefore, atmospheric air gasification of biomass for FT synthesis is preferable.

For the base case without tail gas re-circulation, the conversion of biomass carbon into liquid biofuels remains significantly low for the different FT CO-conversion efficiencies. Significant part of the biomass carbon remains in the tail gas in the form of CO and light hydrocarbons C1-C4 (accounting for 6.50% up to 19.50% of the carbon if FT CO-conversion is 80% down to 40% respectively).

4.2 Re-using Tail Gas to Increase Biofuel Production.

Re-circulation of the tail gas carbon content to the gasifier can increase the productivity of liquid biofuels. This can only be feasible without any further energy consumption for the case of atmospheric gasification due to the higher pressure of the tail gas.

Nevertheless problems would arise from the re-circulation of the inert N_2 and CO_2. But since the tail gas pressure is at least 20 bar, membrane separation techniques could be applicable for the separation of CO, H_2 and C1-C4. Experimentally, membranes composed of aligned carbon nanotubes embedded in a silicon nitride matrix and in a polymer matrix have been reported to exhibit rapid transport of these component gases (Holt et al., 2006; Hinds et al., 2004). Additionally, carbon nanotubes are excellent sorbents for CO and light hydrocarbons, therefore high separation efficiency is feasible (Andrews, 2001).

Table 3. Efficiencies and Biomass to Liquid Conversions

	Atmospheric Gasification FT CO-conversion efficiency 80%	
η_{tot}	η_{biof}	η_{el}
36.26%	28.71%	7.55%
η_{ex}	$\eta_{ex,biof}$	$\eta_{ex,el}$
34.26%	27.04%	7.22%
BtL (kg liquid / kg dry biomass)		11.30%
Biomass C-conversion		25.94%
	Pressurised Gasification FT CO-conversion efficiency 80%	
η_{tot}	η_{biof}	η_{el}
32.31%	17.36%	14.31%
η_{ex}	$\eta_{ex,biof}$	$\eta_{ex,el}$
30.64%	16.36%	14.28%
BtL (kg liquid /kg dry biomass)		6.83%
Biomass C-conversion		16.27%
	Atmospheric Gasification FT CO-conversion efficiency 60%	
η_{tot}	η_{biof}	η_{el}
30.92%	21.52%	9.40%
η_{ex}	$\eta_{ex,biof}$	$\eta_{ex,el}$
29.26%	20.28%	8.98%
BtL (kg liquid /kg dry biomass)		8.48%
Biomass C-conversion		19.46%
	Atmospheric Gasification FT CO-conversion efficiency 40%	
η_{tot}	η_{biof}	η_{el}
25.58%	14.35%	11.23%
η_{ex}	$\eta_{ex,biof}$	$\eta_{ex,el}$
24.26%	13.52%	10.73%
BtL (kg liquid /kg dry biomass)		5.65%
Biomass C-conversion		12.97%

The exergetic efficiency of the membrane separation itself is around 100%, and the only minor exergetic losses are accounted due to the pressure losses of the permeate stream. Exergy losses of the gas compression are not accounted here since this step is anyway required for the FT. A recirculation ratio of CO, H_2 and C1-C4 is defined with Eq. (12). Figure 7 illustrates the effect of *Rec* to the conversion of the carbon to liquid biofuels, for different FT CO-conversion efficiencies. Its increase acts positively on the overall conversion of carbon and allows less intensive FT reactors to be employed and at the same time maintaining high biofuel productivity.

$$Rec = \frac{(\dot{N}_{CO} + \dot{N}_{H2} + \sum_{i=1}^{4} \dot{N}_{Ci})_{\text{Recirculate}}}{(\dot{N}_{CO} + \dot{N}_{H2} + \sum_{i=1}^{4} \dot{N}_{Ci})_{in \text{ the FT Product}}} \quad (12)$$

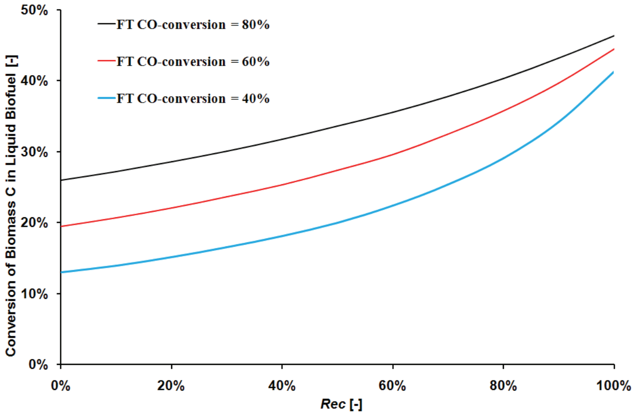

Figure 7. Effect of Rec on the Conversion of the carbon Contained in Biomass to Liquid Biofuels.

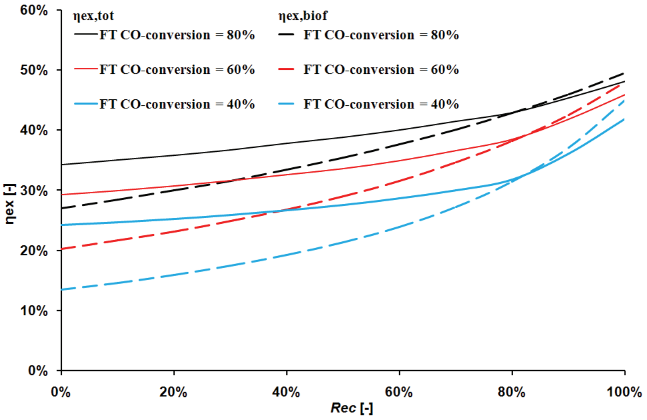

Figure 8. Effect of Rec on the Exergy Efficiency of the System.

Figure 8 shows the effect of *Rec* on the biofuel production exergy efficiency as well as on the total efficiency (including power production) when employing FT CO-conversion from 40-80%. High biofuel production efficiencies can be achieved with FT-reactor operating at even 40% CO-conversion, while the net power production of the system is significantly cut down, due to the gradual reduction of the available exergy towards the Rankine cycle. Values of *Rec* >80% result in shortage of power production to cover the self consumption requirements of the overall process.

5. Conclusions

A detailed process modelling of an air blown atmospheric pressure gasifier integrated with FT synthesis and a Rankine cycle for power production was accomplished. Such process design allows the re-circulation part of FT-tail gas back into the process. Inert constituents such as N_2 and CO_2, would then have to be removed via membrane separation techniques. Overall, the system's own consumption of power can be covered by the power produced using recuperated heat from product gas cooling, and superheating via remaining FT tail gas combustion. The recirculation of tail gas can increase the exergetic efficiency from 36.25% up to 49.47%.

Nomenclature

E	Total exergy of a material stream (W)
E_T^Q	Thermal exergy of heat available at T (W)
E^W	Work or power (W)
h	Enthalpy of a stream (W)
h_o	Standard enthalpy at (To, po) (W)
h_{fg}	Water vaporization enthalpy of 2,442 kJ kg^{-1}
I	Irreversibility (W)
LHV	Lower Heating Value for solids (J kg^{-1})
$\dot{N}$	Mole flow rate (mol s^{-1})
P_i	Partial pressure of component i (bar)
Q	Heat stream (W)
R	8.314 J mol^{-1} K^{-1}; Ideal gas constant
s	Entropy of a stream (W)
s_o	Standard entropy of a stream (kW)
T	Temperature (K)
w	Moisture mass fraction in fuel, (w/w)
W_n	Product n_C weight fraction
x_i	Mole fraction of component i
Greek	
α	probability of chain growth
ε_{oi}	Standard molar chemical exergy of a component i in an ideal mixture (J mol^{-1})
ε_{ph}	Specific physical exergy (J mol^{-1})
ε_{ch}	Specific chemical exergy of ideal stream (J mol^{-1})
$\varepsilon_{ch,biom}$	Specific chemical exergy of biomass (J mol^{-1})
η	efficiency
Subscripts	
biof	Biofuel
el	Electrical
ex	Exergetic
Gas	Gaseous stream (ideal)
gas	Gasification
gccs	Gas Cleaning & Conditioning System
Rec	Recirculation
tot	total
IN	input streams to a control volume
OUT	outgoing streams from a control volume

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