Thermo-economic optimisation of the integration of electrolysis in SNG production from wood

Martin Gassner, François Maréchal

Laboratory for Industrial Energy Systems
Ecole Polytechnique Fédérale de Lausanne
CH – 1015 Lausanne, Switzerland

Abstract

Converting wood to grid quality methane allows to distribute a CO_2 free, renewable energy resource in a conventional energy distribution system and use it in transportation applications. Applying a multi-objective optimisation algorithm to a previously developed thermo-economic process model for the thermochemical production of synthetic natural gas from wood, the present paper assesses the prospect of integrating an electrolyser in conversion systems based on directly and indirectly heated gasification. Due to an inherent lack of hydrogen for complete conversion of wood into methane and the possibility for rational use of oxygen, it is shown that electrolysis is an efficient and economically interesting option for increasing the gas output of the process while storing electricity and producing fuel that mitigates CO_2 emissions.

Keywords: process design, thermo-economic modelling, optimisation, biofuels, SNG

Nomenclature

Roman letters

c	concentration	%vol
C_{GR}	Grass roots cost	k€
C_{OP}	Operating costs	€/MWh
C_P	Total production costs	€/MWh
$e_{a,i}$	Specific avoided CO_2 emissions assigned to substance i	kg/MWh
$e_{p,i}$	Specific CO_2 emissions assigned to the production of substance i	kg/MWh
$e_{u,i}$	Specific CO_2 emissions assigned to the usage of substance i	kg/MWh
$\dot{m}$	Mass flow	kg/s
p_g	Gasification pressure	bar
p_m	Methanation pressure	bar
$p_{s,p}$	Steam production pressure	bar
r_{H2}	Additional hydrogen for methanation	%wt
s_i	Stoichiometric coefficient of substance i	-
SN	Stoichiometric number	-
SNG	Synthetic natural gas	
T_d	Drying temperature	°C
$T_{g,p}$	Preheat temperature of gasification agent	°C
T_g	Gasification temperature	°C
T_m	Methanation temperature	°C
$T_{s,b2}$	Temperature of bleeding level i	°C
$T_{s,s}$	Steam superheat temperature	°C

| w^+ | Consumed electrical power (entering the system) | kW |
| w^- | Produced electrical power (leaving the system) | kW |

Greek letters

Δh_i^0	Lower heating value of substance i	MJ/kg
ΔH_r^0	Standard heat of reaction	kJ/mol
Δh_{vap}	Latent heat of vaporisation	MJ/kg
Δk_i^0	Exergy value of substance i	MJ/kg
ε	Energy efficiency	%
η	Exergy efficiency	%
μ	Mean value	
Φ_w	Wood humidity	%wt
ρ	Correlation coefficient	-
σ	Standard deviation	

1 Introduction

Thermochemical production of synthetic natural gas (SNG) from wood by means of wood gasification, methane synthesis and further CO_2 removal from the produced gases offers several interesting features. A widely available and renewable energy source which is not in competition with food production is used in a process that may be operated as a CO_2 sink if CO_2 sequestration is realised. Clean gaseous fuel is produced and distributed in existing networks. Its use in transportation applications would allow to cut greenhouse gas emissions in a domain where few solutions for mitigating CO_2 emissions and using lignocellulosic biomass exist.

Currently, different process designs are under investigation (1; 2). From the atomic composition of wood, they all have in common that the gas produced by gasification lacks of hydrogen for completely reforming the carbon into methane, which results in a by-production of CO_2. Furthermore, the overall process is exothermic and cogeneration of electricity is therefore possible. For this reason, integrating an electrolyser in the system would allow to increase the methane yield by adding hydrogen to the carbon flow and to produce pure oxygen that is needed for the gasification process while using the cogenerated power. Alternatively, by importing renewable electricity, the process is a way of storing green electrical power in the form of SNG.

Using the process modelling approach described in (3) and further developed in (1), this paper aims to analyse the integration of electrolysis and its impact on the process design and performances.

2 Process description

2.1 Block flow diagram

A general block flow diagram of the process superstructure with the investigated technological alternatives is shown in Figure 1. In a first process step, chipped wood with properties described in Table 1 is dried to avoid severe exergy losses and enhance the formation of CH_4 during gasification. Indirectly heated, steam blown gasification in an internally circulating fluidised bed usually operating at around 850°C and atmospheric pressure and directly heated, oxygen blown pressurised fluidised bed gasification operating at around 800°C are considered as gasification technologies (5; 6). The oxygen necessary for the latter might be supplied externally or produced on-site using either ion transfer membranes (7) or electrolysis. After gasification, the obtained gases need to be cleaned from impurities to prevent methanation catalyst damage. Being rich in H_2, CO and CO_2, the gas is reformed at around 300 to 400°C in an internally cooled, pressurised fluidised bed reactor where a sufficient amount of steam is added to avoid carbon deposition (8). The synthesis gas is dried and CO_2 is removed in order to increase its calorific
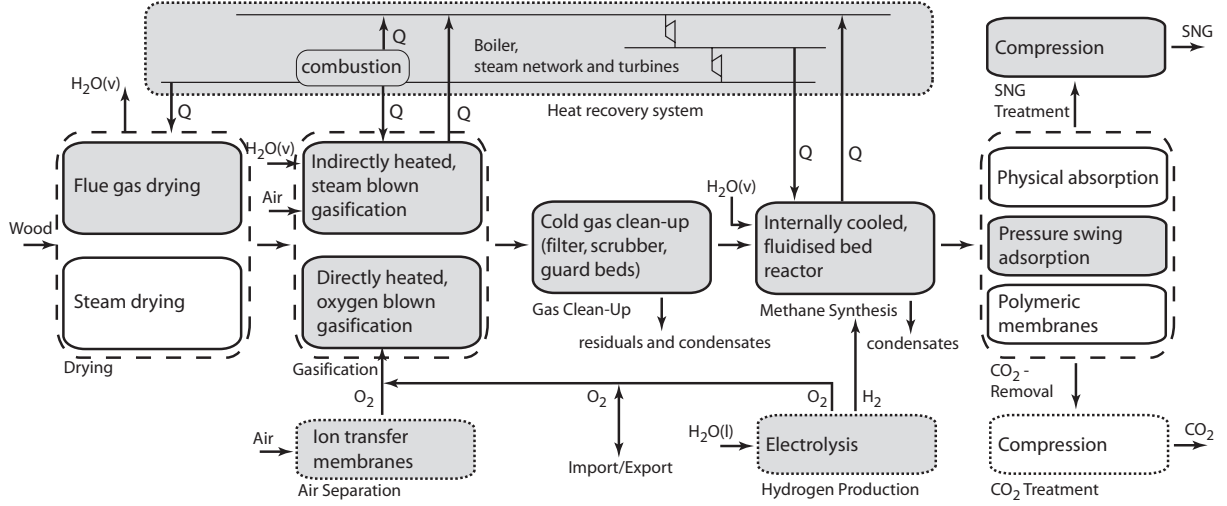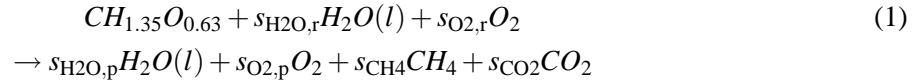
Figure 1: Process superstructure. Dashed boxes assemble competing technologies and dotted ones are used for optional equipment. The process configurations examined in this paper is shown shaded.

value and meet the condition of a Wobbe index between 13.3 and 15.7 kWh/Nm^3 specified for the injection into the gas grid. As excess heat is available from methanation and the flue gases from gasification, the integration of a steam Rankine cycle is used to convert this heat into useful electrical power.

Representing wood as a typical molecule with the carbon atom as reference, the overall conversion that is feasible with the block flow superstructure of Figure 1 is expressed by the following stoichiometry:

$$CH_{1.35}O_{0.63} + s_{\text{H2O,r}}H_2O(l) + s_{\text{O2,r}}O_2 \tag{1}$$
$$\rightarrow s_{\text{H2O,p}}H_2O(l) + s_{\text{O2,p}}O_2 + s_{\text{CH4}}CH_4 + s_{\text{CO2}}CO_2$$

The stoichiometric coefficients s_i depend on the considered technological production route. Two exemplary cases are shown in Table 2. For processes without oxygen imports or exports, the enthalpy of reaction ΔH_r^0 is slightly negative and the conversion is exothermal. If the aim is to completely reform the carbon contained in wood into methane, the ΔH_r^0 becomes positive and indicates an important lack of energy due to water evaporation and separation.

2.2 Integration of electrolysis in the process

The prospect of integrating electrolysis in the process rests upon the lack of hydrogen to completely reform the wood's carbon into methane. Hydrogen might be conveniently added to the methanation,

Table 1: Proximate and ultimate analysis of wood.

Proximate analysis			Ultimate analysis					
Δh_{wood}^0 [a]	16.2	MJ/kg$_{dry}$	C	51.09	%wt	O	42.97	%wt
Δk_{wood}^0 [b]	20.9	MJ/kg$_{dry}$	H	5.75	%wt	N	0.19	%wt
Φ_w [c]	50.0	%wt						

[a] Δh_{wood}^0 is calculated considering the latent heat of vaporisation for moisture and referred to the dry mass of wood, i.e. $\Delta h_{wood}^0 = \Delta h_{wood,dry}^0 - \Delta h_{vap}\Phi_w/(1-\Phi_w)$.

[b] Chemical exergy is calculated according to (4).

[c] Wood moisture is defined on total mass basis, i.e. the water content per mass of wet wood.

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Table 2: Stoichiometric coefficients for two exemplary cases of Equation 1.

	no import or export of O_2	complete reforming of $CH_{1.35}O_{0.63}$ to CH_4
$s_{H2O,r}$	0.3475	1.955
$s_{O2,r}$	0	0
s_{H2}	0.3475	1.955
$s_{O2,g}$	0.17375	0
$s_{H2O,p}$	0	0.63
$s_{O2,p}$	0	0.9775
s_{CH4}	0.51125	1
s_{CO2}	0.48875	0
ΔH_r^0	-10.5 kJ/mol$_{wood}$	425 kJ/mol$_{wood}$

where the following reactions take place:

$$CO + 3H_2 \rightleftharpoons CH_4 + H_2O \qquad \Delta H_r^0 = -206 kJ/mol \qquad (2)$$

$$C_2H_4 + 2H_2O \rightleftharpoons 2CO + 4H_2 \qquad \Delta H_r^0 = 209 kJ/mol \qquad (3)$$

$$CO_2 + H_2 \rightleftharpoons CO + H_2O \qquad \Delta H_r^0 = 41 kJ/mol \qquad (4)$$

The stoichiometric coefficients of these reactions allow to determine the amount of hydrogen that is needed to prevent the formation of CO_2. Considering ethene to represent the higher hydrocarbons, it is possible to define the stoichiometric number SN of the reactant mixture that characterise the methane stoichiometry:

$$SN = \frac{c_{H2}}{3c_{CO} + 4c_{CO2} + 2c_{C2H4}} \qquad (5)$$

To obtain a pure methane stream from the stoichiometry, this ratio must be equal to unity, whereas it is generally lower for gas produced from biomass.

Apart from hydrogen feeding to the methane synthesis section, electrolysis supplies oxygen to the directly heated gasifier. In indirectly heated gasification, the oxygen is used to enrich the combustion air and thus decreases the fuel demand since the reduction of nitrogen dilution increases the amount of heat available at high temperature. The atomic pathways of hydrogen and oxygen that are dissociated during electrolysis are shown in Figure 2.

In this paper, it is assumed that hydrogen is produced in the electrolyser at a temperature of 120°C and an efficiency of 85%, defined as the ratio between the lower heating value of the produced H_2 and the power input. According to the US/DOE target by 2010, capital costs of electrolysers are assumed to be of 300 €/kW$_{el,installed}$ (9).
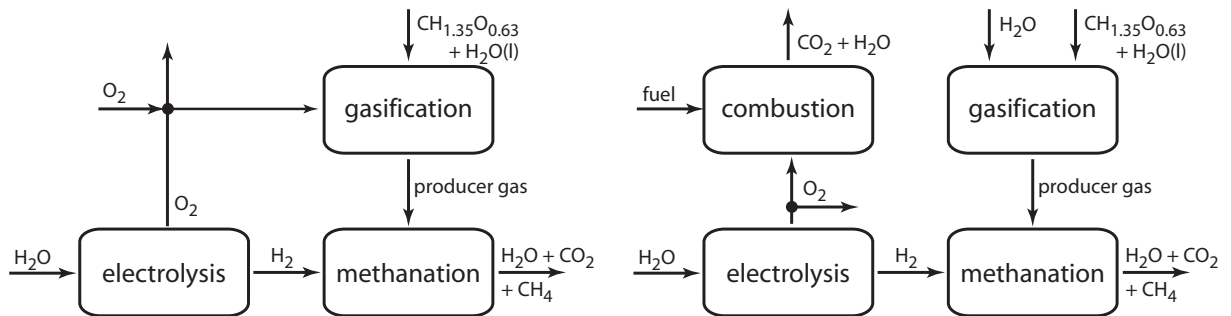

Figure 2: Pathways of hydrogen and oxygen from electrolysis through the directly (left) and indirectly heated systems.

Table 3: Assumptions for the economic analysis.

Parameter	Value	Parameter	Value
Marshall&Swift index (2004)	1197	Operators	4 p./shift
Dollar exchange rate	1 €/US$	Operator salary	60 k€/year
Interest rate	6%	Oxygen price	70 €/ton
Expected lifetime	15 years	Wood price (Φ_w=50%wt)	16.7 €/MWh
Plant availability	90%	Electricity price (import)	88.9 €/MWh
Maintenance costs	5%/year of C_{GR}	Electricity price (export)	26.4 €/MWh

3 Thermo-economic modelling

In previous work, a thermo-economic process model considering the different technological options of the superstructure has been developed and reconciled with experimental data (3; 1; 10). Its thermodynamic part consists of an energy-flow and an energy-integration model. The energy-flow model computes the transformation of species and the associated heat requirements using the commercial flowsheeting software *Belsim-Vali 4* (11). This data is implemented in the energy-integration model which determines the optimal thermal process integration and computes the combined heat and power production with the pinch methodology. This allows one to optimise the heat recovery and power production in the Rankine cycle. Considering the thermodynamic conditions as decision variables, the economic model then calculates the capital costs of the plant by sizing the major equipment necessary to reach the process requirements. For this purpose, design rules and cost correlations from (12) have been calibrated on plant data to ensure a valid assessment of the overall investment costs.

The thermodynamic process performances are determined in terms of energy and exergy efficiency, defined by eq. 6 and 7 respectively:

$$\varepsilon = \frac{\Delta h_{SNG}^0 \dot{m}_{SNG} + w^-}{\Delta h_{wood}^0 \dot{m}_{wood} + w^+} \tag{6}$$

$$\eta = \frac{\Delta k_{SNG}^0 \dot{m}_{SNG} + \Delta k_{O2}^0 \dot{m}_{O2} + w^-}{\Delta k_{wood}^0 \dot{m}_{wood} + w^+} \tag{7}$$

In these equations, Δh^0 and Δk^0 designate the lower heating and exergy value per unit mass respectively. w^- refers to overall produced power and w^+ to overall consumed power, whereas only one of these terms occurs in the equations since the overall power balance is of interest. The economic performances of the process are determined as outlined in (1) using the assumptions of Table 3. The capital costs of the plant are denominated grass roots cost C_{GR} and correspond to the initial investment without land. The operating costs C_{OP} include estimated expenses for raw materials, utilities, labour and maintenance for the production of SNG and account also for the benefit from the sale of surplus oxygen from the electrolyser. Total production costs C_P designate the expected cost of manufacturing including the investment's depreciation. The environmental impact of the process is assessed with a CO_2 emission inventory which includes the avoided emissions due to the substitution of the natural gas consumption from the grid.

4 Impact of electrolysis

4.1 Process optimisation strategy

The impact of electrolysis on the process design and performances has been investigated by applying an evolutionary, multi-objective optimisation algorithm (13) to the process layouts that are shown shaded on Figure 1. The wood to methane conversion is based on flue gas wood drying, directly or indirectly heated gasification, methanation and pressure swing adsorption for SNG purification. The high temperature energy balance is satisfied by burning process waste streams and produced syngas when needed. Excess

Table 4: Decision variables for optimisation.

Section	Variable	Variation domain		Section	Variable	Variation domain	
Drying	$T_{d,in}$	[160; 240]	°C	Methane synthesis	p_m	[1; 50]	bar
	$\Phi_{w,out}$	[5; 35]	%wt		$T_{m,in}$	[300; 400]	°C
Gasification	p_g	[1; 20]	bar		$T_{m,out}$	[300; 400]	°C
	T_g	[800; 900][a]	°C		r_{H2}[c]	[0; 15]	%wt
		[750; 850][b]	°C	Steam network	$p_{s,p}$	[40; 100]	bar
	$T_{g,p}$	[300; 600]	°C		$T_{s,s}$	[350; 550]	°C
					$T_{s,b2}$	[50; 250]	°C

[a] Indirectly heated gasification.

[b] Directly heated gasification.

[c] r_{H2} is the amount of hydrogen produced by electrolysis that is added to the methanation (in $kg_{H2,added}/kg_{g,out}$).

heat recovery is done through a steam cycle that realises combined heat and power production. The list of decision variables and their domain is given in Table 4. In order to entirely cover the plant performances in terms of energy and exergy efficiency, investment cost and operating costs, the different terms of these indicators are used as objective functions. This allows one to limit the number of objective functions for the optimisation problem to three, i.e. the SNG output, the amount of produced or consumed electricity and the grass roots cost of the plant. For all the calculations, the plant's nominal power based on wood input at 50%wt humidity has been fixed to 20 MW_{th}.

4.2   Optimisation results

Although the optimisation is performed using three objectives, it is more convenient to present the results in terms of the exergy efficiency and the total production costs. This explains why the resulting Pareto plot (3) contains a branch in the suboptimal domain that would not appear if these two indicators had been directly used as objectives in the optimisation. With the aim of comparing and analysing the optimisation results, the optimal configurations are plotted with the parametric addition of hydrogen to a plant designed for operation without electrolysis (dash-dotted line). The process configurations and performances of the designs for minimum production costs, maximum energy efficiency, exergy efficiency and SNG production are detailed in Table 5.

Independently of the chosen gasification technology, the plots show that the process benefits from electrolysis with regard to exergy efficiency, whose maximum values are of 67.1% and 70.6% for directly and indirectly heated gasification respectively. This corresponds to an increase by 2.0% to 3.4% compared to the configurations without an electrolyser. However, due to the high price of electricity, increasing the amount of additional hydrogen increases the specific production costs of SNG considerably. For the given economic environment, the production of SNG at the optimal exergy efficiency is by 11% to 27% more expensive than the minimum cost of 57.1 €/MWh$_{SNG}$ and 56.3 €/MWh$_{SNG}$ for plants based on directly and indirectly heated gasification. The influence of the electrolysis rate on the optimal design of the process as well as its efficiency, cost and environmental impact are discussed in detail in sections 4.3 to 4.6.

4.3   Impact of electrolysis on process design

Dashed lines on Figure 3 show the evolution of efficiency and total production costs if hydrogen from electrolysis is continuously added to the process without changing its design. For increasing rates of electrolysis, these solutions are clearly suboptimal compared to the optimised configurations, which demonstrates that the integration of the usage of an electrolyser modifies the design of the whole process. In order to get an idea of this impact, the correlation coefficients ρ between the decision variables and
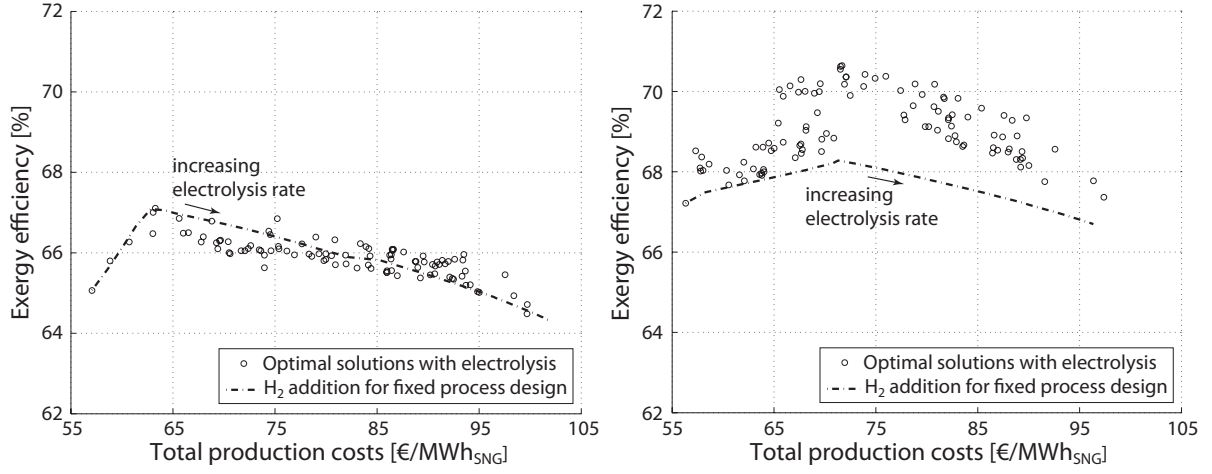
Figure 3: Optimal trade-off for directly (left) and indirectly heated gasification after 25'000 iterations.

Table 5: Configurations and performances of some selected process designs.

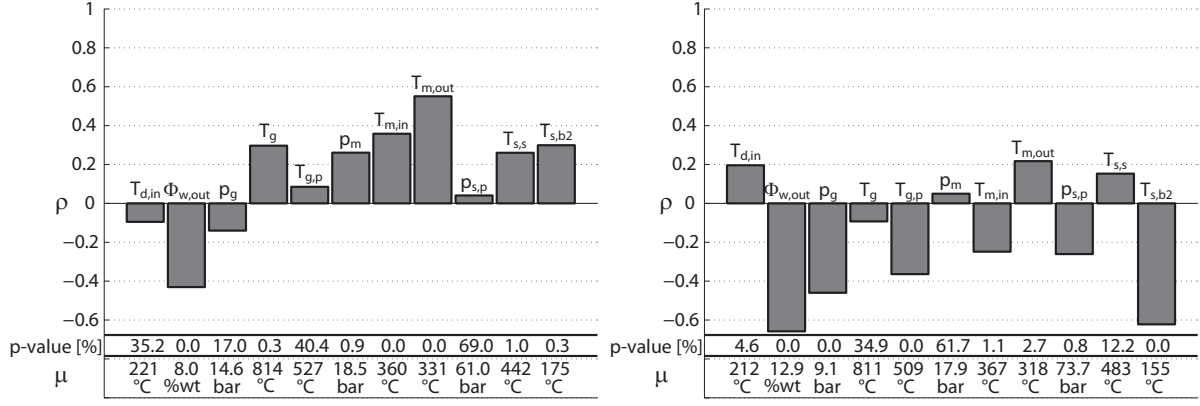|	Unit	directly heated gasification				indirectly heated gasification			
		$C_{P,min}$	ε_{max}	η_{max}	SNG_{max}	$C_{P,min}$	ε_{max}	η_{max}	SNG_{max}
$T_{d,in}$	°C	236	239	235	212	228	197	230	231
$\Phi_{w,out}$	%wt	6.04	8.1	6.03	6.86	22.9	22.3	6.95	7.91
p_g	bar	16.7	15.1	16.7	16.3	13.8	6.94	8.39	7.62
T_g	°C	804	801	804	805	828	805	809	819
$T_{g,p}$	°C	527	514	526	505	520	509	459	443
p_m	bar	28.9	19.7	28.9	32.8	10.7	13.0	16.6	21.2
$T_{m,in}$	°C	355	338	356	361	368	379	364	387
$T_{m,out}$	°C	343	302	343	327	305	311	362	374
r_{H2}	%wt	0.00	0.24	1.26	12.7	0.00	0.22	3.36	13.0
$p_{s,s}$	°C	57.3	65.6	57.1	65.7	74.7	99.3	80.9	62.0
$T_{s,s}$	°C	350	384	361	411	473	464	474	499
$T_{s,b2}$	bar	161	53.2	161	188	238	246	166	165
w^+	MW	0.35	0.96	3.22	28.9	0.64	1.17	7.66	28.4
$\Delta h^0_{wood}\dot{m}_{wood}$	MW	20.0	20.0	20.0	20.0	20.0	20.0	20.0	20.0
$\Delta k^0_{wood}\dot{m}_{wood}$	MW	25.8	25.8	25.8	25.8	25.8	25.8	25.8	25.8
$\Delta h^0_{SNG}\dot{m}_{SNG}$	MW	16.4	17.0	18.8	34.0	17.2	17.8	22.8	35.4
$\Delta k^0_{SNG}\dot{m}_{SNG}$	MW	17.0	17.6	19.5	35.3	17.8	18.5	23.7	36.7
$\Delta k^0_{O2}\dot{m}_{O2}$	kW	0.00	0.00	0.00	119	0.00	0.00	8.07	108
ε	%	80.7	81.1	81.0	69.6	83.2	84.3	82.5	73.1
η	%	65.1	65.8	67.1	64.5	67.2	68.5	70.6	67.6
C_{GR}	M€	21.6	21.9	21.3	36.6	21.7	22.1	24.5	38.4
C_{OP}	€/MWh$_{SNG}$	39.5	41.6	48.1	85.3	39.4	40.7	57.3	81.8
C_P	€/MWh$_{SNG}$	57.1	58.8	63.3	99.7	56.3	57.3	71.7	96.4

Figure 4: Correlation of decision variables and ṙ$_{H2}$ for directly (left) and indirectly heated gasification.

the amount of additional hydrogen r_{H2} produced by electrolysis have been computed by Equation 8.

$$\rho_{i,r_{H2}} = \frac{\mu\left(x_i - \mu(x_i)\right)\mu\left(r_{H2} - \mu(r_{H2})\right)}{\sigma(x_i)\sigma(r_{H2})} \tag{8}$$

In this equation, x_i designates the observations of the set i, μ the corresponding mean values and σ their standard deviation. Positive values of these coefficients indicate that an increase of the value is expected when the hydrogen flowrate increases.

Figure 4 shows the correlation coefficient and the according p-value quantifying the probability that such a correlation is obtained from random data. For directly heated gasification, significant interdependences between the electrolysis rate and the other decision variables are observed for the extent of wood drying, the gasification temperature, the operating conditions of methane synthesis and the steam cycle temperatures. These variables are in strong relation to the thermal integration of the conversion of hydrogen into methane. Since increasing the amount of additional hydrogen increases the heat available from the exothermal reaction, a higher methanation outlet temperature allows transferring heat with higher exergetic value to the steam cycle. Furthermore, more excess heat can be used for wood drying to cut the losses in the gasifier due to water evaporation. The latter is observed even more pronounced in case of steam blown gasification. In this case however, no general trend can be derived from the temperature changes of the stream and a more detailed analysis of the process integration is necessary.

The influence of electrolysis on the process sections gets apparent by investigating its correlation with the investment costs (Figure 5). Due to the increased amount of reformed methane, larger equipments for its synthesis, the associated power production and the overall heat exchange are needed, while the presence of less CO_2 in the product gas decreases the expenses for its removal. Especially in case of indirectly heated gasification, the process sections upstream of the methane synthesis are indirectly affected by the need to dry wood more extensively and due to the lower gasification pressure, which causes itself decreasing expenses for the gasifier and increasing costs for gas cleaning.

4.4 Impact of electrolysis on process efficiency

Figure 6 shows the evolution of the overall plant efficiencies as a function of the relative input power to the electrolyser. In directly heated gasification, both the energy and exergy efficiency increase for low rates of electrolysis and reach maxima of 81.1% and 67.1% at relative electrolyser capacities of 0.03 MW$_{el}$/MW$_{wood}$ and 0.14 MW$_{el}$/MW$_{wood}$ respectively. For the parametric H$_2$ addition data, the maximum values of both efficiencies occur at the same electrolysis rate that is just sufficient to satisfy the oxygen demand of the gasifier. If the rate is increased further, oxygen is produced in excess and the energy efficiency in particular decreases significantly. In indirectly heated gasification, the efficiencies are higher than in the directly heated case and an optimal energy efficiency of 84.3% is observed at a low electrolyser power of 0.02 MW$_{el}$/MW$_{wood}$. The exergy efficiency reaches its maximum value of 70.6% at
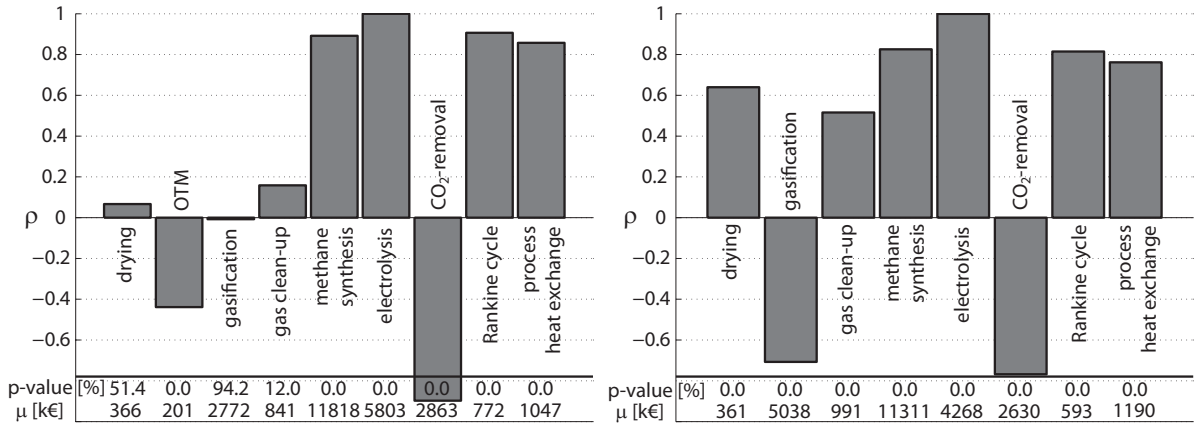
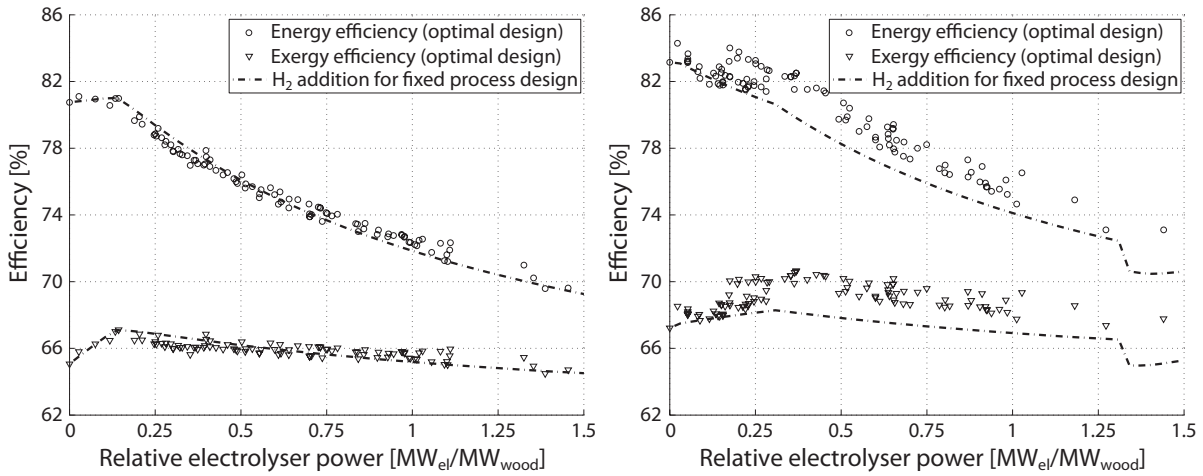Figure 5: Correlation of equipment costs and $\dot{n}_{H_2}$ for directly (left) and indirectly heated gasification.

Figure 6: Impact of electrolysis on efficiency for directly (left) and indirectly heated gasification.

$0.37\ \mathrm{MW_{el}/MW_{wood}}$. In case of parametric addition of hydrogen, an exergetic optimum is again observed at the point where a maximum amount of oxygen is used on-site, yet without any excess.

Oxygen blown gasification benefits thus more significantly from the use of an electrolyser due to the lower initial efficiency and more striking advantages from substituting the oxygen production by means of ion transfer membranes. In indirectly heated gasification, the overall process efficiency of an optimised plant without additional hydrogen reaches almost the level of the electrolyser itself. The fuel savings achieved through oxygen enriched combustion do not balance the losses induced by processing the additional hydrogen. The benefits are only observed in terms of exergy efficiency.

4.5 Impact of electrolysis on process economics

4.5.1 Specific production costs

The impact of electrolysis on the specific production costs of synthetic natural gas are shown in Figure 7. Breakdowns of the total production costs into investment and operating costs of electrolysis and the other process sections are shown in Table 6. In order to highlight the influence of the capital costs for electrolysers, production costs considering investment costs of 300 and 1000 €/kW$_{el,installed}$ are plotted. While the former represents a targeted cost by 2010, the latter is seen as a lower bound for investments up to year 2004 (9).

The current average market price of electricity in Switzerland prevents the continuous addition of hydrogen to be economically viable. In both cases, the specific production costs are considerably increased
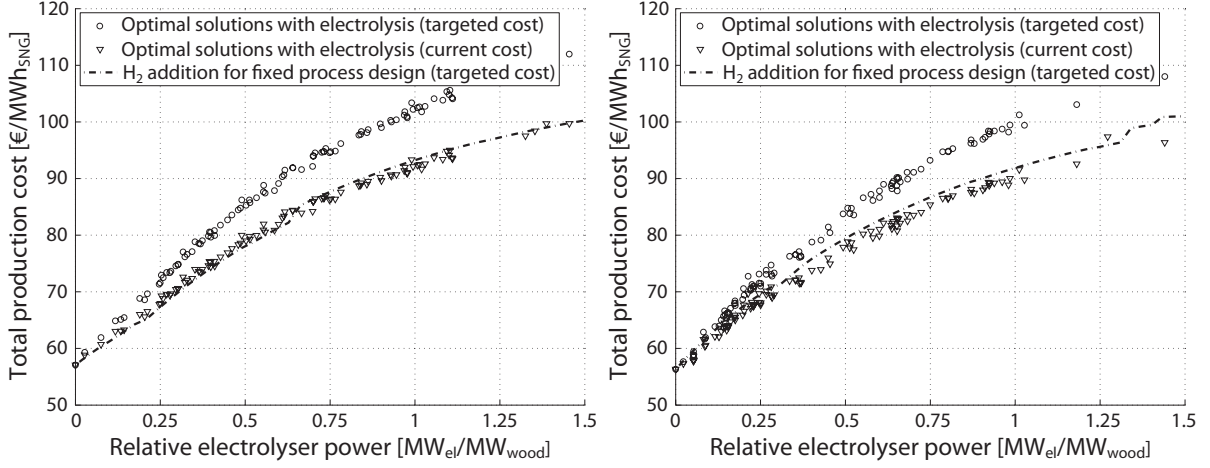
Figure 7: Impact of electrolysis on production costs for directly (left) and indirectly heated gasification for two different investment costs of the electrolyser.

Table 6: Cost breakdowns [€/MWh$_{SNG}$].

Cost type	directly heated gasification				indirectly heated gasification			
	$C_{P,min}$	ε_{max}	η_{max}	SNG_{max}	$C_{P,min}$	ε_{max}	η_{max}	SNG_{max}
$C_{GR,electrolysis}$	0.0	0.2	0.9	5.2	0.0	0.2	2.0	5.0
$C_{GR,process}$	17.6	17.0	14.2	9.2	16.9	16.4	12.4	9.5
$C_{OP,electrolysis}$	0.0	2.9	13.8	77.0	0.0	2.3	29.1	73.2
$C_{OP,process}$	39.5	38.7	34.4	8.3	39.4	38.4	28.2	8.6
C_P	57.1	58.8	63.3	99.7	56.3	57.3	71.7	96.3

with increasing electricity consumption. The break-even price of electricity which equals the cost SNG production without electrolysis is of 38.8 and 40.2 €/MWh$_{el}$ for maximum gas production with directly and indirectly heated gasification respectively.

4.5.2 Process profitability

Although the specific production costs of SNG tend to increase with the addition of hydrogen produced by electrolysis, the profit from treating a fixed amount of wood might increase due to the generation of additional gas. This effect is analysed on Figure 8, where the most profitable process operation depending on electricity cost and gas price is shown. By comparison with typical costs of generating electricity and Swiss gas prices (14; 15), the plots indicate that electrolysis might effectively allow to increase the revenue from wood, if not the market price of electricity, but its real production costs are considered. With decreasing expenses for the consumed electricity, plant designs for optimal exergy efficiency and finally for maximal gas yield from biomass get most profitable. In particular, electricity generated by nuclear power allows for profitable base load operation of electrolysis at gas prices higher than 40 to 45 €/MWh$_{SNG}$, while wind power requires gas prices higher than 50 to 80 €/MWh$_{SNG}$, which corresponds to the range of transportation applications.

4.5.3 Marginal production costs

As outlined in section 2.2, the stoichiometric number of the gas from wood gasification is lower than unity and the gas production is only limited by the available hydrogen. The marginal production of SNG from electricity by means of electrolysis can be estimated by rearranging and differentiating Equation 6:

$$\Delta h^0_{SNG} \frac{\partial \dot{m}_{SNG}}{\partial w^+} = \frac{\partial \varepsilon}{\partial w^+} \left(\Delta h^0_{wood} \dot{m}_{wood} + w^+ \right) + \varepsilon \left(\Delta h^0_{wood} \frac{\partial \dot{m}_{wood}}{\partial w^+} + 1 \right) \tag{9}$$
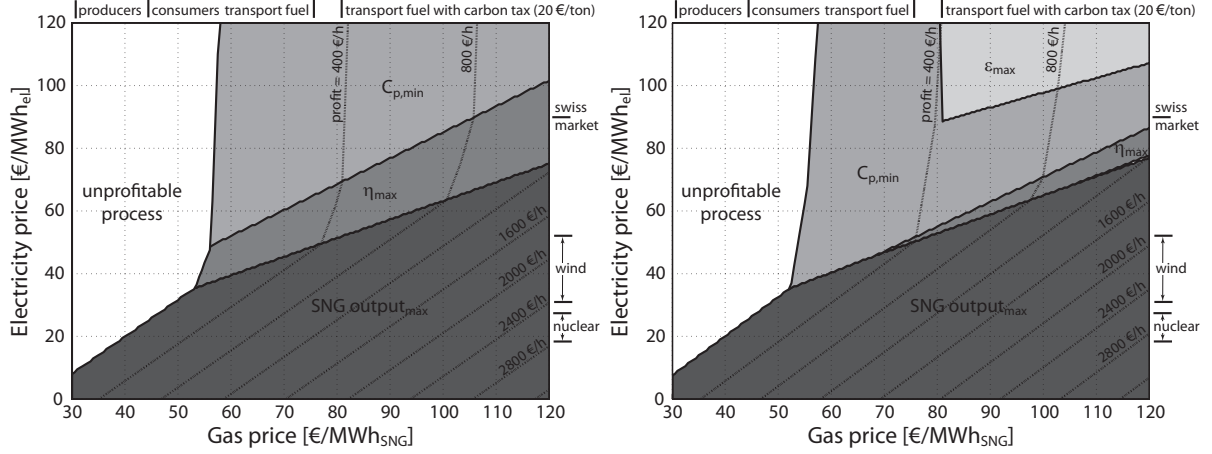
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Figure 8: Design types for maximum profit for directly (left) and indirectly heated gasification.

In marginal conditions, we assume a constant process efficiency ($\partial \varepsilon / \partial w^+ = 0$) and a fixed flow of wood ($\partial \dot{m}_{wood} / \partial w^+ = 0$), from what follows:

$$\Delta h^0_{SNG} \frac{\partial \dot{m}_{SNG}}{\partial w^+} = \varepsilon \qquad (10)$$

It is thus possible to store electricity as natural gas with an efficiency equal to the overall process efficiency and to produce additional SNG at the marginal cost of electricity factored by $1/\varepsilon$. In this way, adding an electrolyser to the system allows to absorb seasonal overproduction of electrical power and efficiently produce fuel for transport applications.

4.6 Impact of electrolysis on CO_2 balance

Based on the data for an emission inventory depicted in Table 7, Table 8 shows the specific avoided emissions of CO_2 due to the substitution of fossil natural gas by synthetic natural gas produced from wood. Values for both the Swiss and UCTE electricity mix as well as electricity generated from renewable sources are given. In addition to the avoided emissions for a process without CO_2 sequestration, values corresponding to an optional carbon dioxide sequestration are further shown.

The impact of adding hydrogen from electrolysis to the process is influenced by different effects. Firstly, the SNG production and hence the amount of substituted natural gas as well as the avoided CO_2 emissions are increased. If the electricity needed for this purpose is generated from fossil sources, this is counterbalanced by additional emissions of carbon dioxide associated with the power demand. Furthermore, adding hydrogen to the carbon flow decreases the ratio of CO_2 emitted on-site to CO_2 emitted during combustion of SNG. If carbon dioxide is not captured at the process outlet, this does however not change the total emissions of carbon originated from wood. The only effect on the overall CO_2 balance is that the emissions of fossil natural gas are substituted by the ones for electricity production. Reminding

Table 7: Data for CO_2 emission inventory (16).

Type	Associated emissions		Type	Associated emissions	
Wood growth	-418	kg_{CO2}/MWh_{wood}	Swiss electricity ($e_{p,el}$)	110	kg_{CO2}/MWh_{el}
Wood chopping[a]	5.38	kg_{CO2}/MWh_{wood}	UCTE electricity ($e_{p,el}$)	450	kg_{CO2}/MWh_{el}
Wood transport[b]	0.87	kg_{CO2}/MWh_{wood}	NG production ($e_{p,NG}$)	26.7	kg_{CO2}/MWh_{NG}
			NG combustion ($e_{u,NG}$)	203	$kg_{CO2}/MWh_{(S)NG}$

[a] data for Φ_w=55%wt
[b] average distance of 40 km with lorry (16t)

Table 8: Linear regressions for avoided fossil CO_2 emissions [kg_{CO2}/MWh_{wood}] as a function of electricity input [MW_{el}] to the electrolyser: $e_{a,el} = c_1 + c_2 w^+_{electrolysis}$.

	Gasification	with sequestration			without sequestration		
		green	CH-mix	UCTE-mix	green	CH-mix	UCTE-mix
c_1	directly heated	434	432	427	187	186	182
	indirectly heated	435	432	422	196	193	184
c_2	directly heated	0.78	-4.53	-20.9	6.76	1.45	-11.9
	indirectly heated	0.85	-4.49	-21.0	7.31	1.97	-14.5

Equation 10, it is possible to produce SNG from electrical power at an efficiency equal to the process efficiency. Accordingly, the overall greenhouse gas emissions decrease if

$$e_{p,el} < \varepsilon \cdot e_{u,NG} \approx 188 \, kg_{CO2}/MWh_{el} \tag{11}$$

and specific avoided emissions of

$$e_{a,el} = \varepsilon \cdot e_{u,NG} - e_{p,el} \tag{12}$$

are assigned to the electricity used in the process. If carbon dioxide is captured at the process outlet, adding hydrogen results in a smaller amount of CO_2 that is sequestrated, but emitted during combustion of the additionally produced SNG. Only the emissions related with the production and transportation of natural gas are mitigated and a decrease of the total emissions is obtained if

$$e_{p,el} < \varepsilon \cdot e_{p,NG} \approx 21.8 \, kg_{CO2}/MWh_{el} \tag{13}$$

resulting in specific avoided emissions of

$$e_{a,el} = \varepsilon \cdot e_{p,NG} - e_{p,el} \tag{14}$$

CO_2 sequestration will therefore require an electricity production based on renewable resources.

5 Conclusion

Exploring a detailed process model of thermochemical production of SNG from wood and using a multi-objective optimisation framework, the impact of electrolysis on process design and its thermodynamic, economic and environmental performances have been investigated. It has been shown that the process design is affected by the integration of electrolysis. Due to an appropriate use of hydrogen in the methanation, the integration of an electrolyser allows to increase the energy and exergy efficiency of the plant. However, the actual market price of electricity economically disfavours to compensate the lack of hydrogen in the biomass by the use of electrolysis. Nevertheless, if electricity is available at the cost of its generation or if it is seasonally cheap, electrolysis increases the profit from the conversion of wood and allows to efficiently convert and store electricity in the form of transportation fuel. Furthermore considering the possible CO_2 sequestration and the substitution of fossil natural gas, CO_2 emission are mitigated and negative specific emissions are allocated to the electricity used in the process, especially when renewable electricity is available.

References

[1] Gassner, M., Maréchal, F. *Thermo-economic model of a process converting wood to methane.* Submitted to Biomass & Bioenergy, 2006.

[2] Mozaffarian, M., Zwart, R. *Feasibility of biomass/waste-related SNG production technologies.* ECN-C-03-66, Petten, Netherlands, 2003.

[3] Duret, A., Friedli, C., Maréchal, F. *Process design of Synthetic Natural Gas (SNG) production using wood gasification.* Journal of cleaner production 2005, 13(15), 1434-1446.

[4] Szargut, J., Styrylska, T. *Angenäherte Bestimmung der Exergie von Brennstoffen.* BWK 1964, 16 (12), 589-596.

[5] Hofbauer, H. et al. *Six years experience with the FICFB-gasification process.* 12th European conference and technology exhibition on biomass for energy, industry and climate protection, Amsterdam, Netherlands, 2002.

[6] *Gas production/gas treating* Ullmann's encyclopedia of industrial chemistry. Online edition, 2003.

[7] van Stein, E.E., Juwono, E., Demetri, E.P. *The impact of ITM oxygen on economics for coal based IGCC.* 27th International technical conference on coal utilization & fuel systems, Clearwater, Florida, USA, 2002.

[8] Stucki, S. *Projet bois-methane. Rapport sur la clôture de la phase 1 du projet: Preuve de la faisabilité technique à l'échelle du laboratoire.* PSI, Villigen, Switzerland, 2005.

[9] Newborough, M. *A report on electrolysers, future markets and the prospects for ITM Power Ltd's electrolyser technology.* Available at: www.h2fc.com/Newsletter

[10] Gassner, M. *Energy Integration and thermo-economic evaluation of a process converting wood to methane.* Masters Thesis. LENI, Ecole Polytechnique Fédérale de Lausanne, Switzerland, 2005.

[11] www.belsim.com

[12] Ulrich, G.-D. *A guide to chemical engineering process design and economics.* Wiley, New York, 1984.

[13] Leyland, G.B. *Multi-objective optimisation applied to industrial energy problems.* PhD thesis no. 2572, Swiss Federal Institute of Technology, Lausanne, 2002.

[14] *Projected costs of generating electricity. 2005 Update.* OECD/IEA, Paris, 2005.

[15] *Statistique globale suisse de l'énergie 2004.* OFEN, Bern, 2005.

[16] Misc. publications from *ecoinvent - Swiss centre for life cycle inventories.* Available at: www.ecoinvent.ch