



Thermo-economic evaluation and optimization of the thermo-chemical conversion of biomass into methanol



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ABSTRACT

In a carbon and resources constrained world, thermo-chemical conversion of lignocellulosic biomass into fuels and chemicals is regarded as a promising alternative to fossil resources derived products. Methanol is one potential product which can be used for the synthesis of various chemicals or as a fuel in fuel cells and internal combustion engines. This study focuses on the evaluation and optimization of the thermodynamic and economic performance of methanol production from biomass by applying process integration and optimization techniques. Results reveal the importance of the energy integration and in particular of the cogeneration of electricity for the efficient use of biomass.

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1. Introduction

Methanol was produced since the early 1800s through the distillation of wood to make 'wood alcohol'. This method was replaced in the 1920s by large scale processes producing methanol from hydrogen and carbon oxides mixture obtained from the incomplete combustion and reforming of fossil fuels. Today, methanol is produced mainly by reforming of natural gas, naphtha or refinery light gas [1]. Other ways to produce methanol, which are currently being investigated, include direct methane oxidation without the intermediate step of syngas production and reductive hydrogenation recycling of CO₂, requiring hydrogen, but providing a way to use CO₂. The price of methanol is about 16 €₂₀₁₁/GJ [2], while as a reference, the OPEC (Organization of the Petroleum Exporting Countries) basket price of crude oil is 107.46 US\$/bbl, or approximately 14.7 €₂₀₁₁/GJ. The recent peaks in methanol prices at the end of 2007 and 2008 are mainly due to plants shutting down for scheduled maintenance but also to the increasing demand in the growing economies [3]. Methanol is mainly used as a feedstock

for the synthesis of other products. Being a fuel with an octane number of 100, it can be blended with gasoline as an oxygenated additive or used directly in internal combustion engines with only minor modifications [3]. Furthermore, methanol can be used to produce electricity in DMFC (direct methanol fuel cells) or DME (dimethyl ether) which can be used as a substitute to diesel fuel as well as household gas for cooking and heating, and gasoline (methanol to gasoline synthesis) [4]. Only a few studies analyzed the technical and economic feasibility of the thermo-chemical conversion of biomass into methanol and explored different process configurations. A comparison of the biomass derived methanol production costs is carried out by Spath and Dayton [5] who reported values varying from 10 to 19.6 US\$₁₉₈₉/GJ_{HHV} (28–54 €₂₀₁₁/GJ_{LHV}, plant capacity 390 MW_{th}) for the study of Wyman et al. [6] to 9–12 US\$/GJ_{HHV} (18–23 €₂₀₁₁/GJ_{LHV}, plant capacity 410 MW_{th}) for the study of Hamelinck et al. [7]. Sarkar et al. [8] reported, for Western Canada, 14 to 23 US\$₂₀₀₈/GJ_{HHV} (23–36 €₂₀₁₁/GJ_{LHV}, plant capacity 430 MW_{th}). Hamelinck et al. investigated promising conversion concepts and compared different types of gasifiers and gas cleaning steps obtaining overall HHV (high heating value) energy efficiency of 55%. The models developed by Van Rens et al. [9] and Huisman et al. [10] addressed two process configurations: a present day design relying on proven technologies (though not on commercial scale for biomass applications) and a near future design studied within the CHRISGAS project [11] in particular for syngas

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Nomenclature

Abbreviations

AGR	acid gas removal
BM	biomass
DME	dimethyl ether
EF	entrained flow
FICFB	fast internally circulating fluidized bed
FT	Fischer–Tropsch
MeOH	methanol
WGS	water–gas shift
HHV	higher heating value MJ/kg
LHV	lower heating value MJ/kg

Greek letters

ϕ	humidity %
η_{en}	overall energy efficiency %
η_{chem}	chemical efficiency %
$\eta_{\text{en_eq}}$	equivalent efficiency %

Roman letters

\dot{E}	mechanical/electrical power kW
\dot{m}	mass flow kg/s
P	pressure bar
T	temperature °C or K

Superscripts

+	material or energy stream entering the system
-	material or energy stream leaving the system

conversion of lignocellulosic biomass into syngas and liquid fuels (FT (Fischer–Tropsch), MeOH (methanol), DME) [13–15]. The analysis is completed here by applying the thermo-economic process optimization methodology described by Gassner et al. [14], focusing on the gasification technology. An EF (entrained flow) gasifier and a FICFB (fast internally circulating fluidized bed) gasifier are compared, as represented in Fig. 1. The superstructure is built of single process-units thermo-economic models that can be assembled to systematically study different process configurations. The thermo-chemical models are developed using commercial flowsheet calculation software Belsim and Belsim-Vali [16] providing the chemical transformation and the heat requirements of the process units. These models are coupled with the economic and the energy integration models. The energy integration model computes the minimum energy requirements of the process using the mass balance between the unit operations and the heat cascade as constraints. If the combustion of the waste streams is not sufficient to provide the heat requirement above the pinch, selected process streams may be used as fuels. Available excess heat can be recovered in a Rankine cycle producing electricity. The energy integration model is detailed in Refs. [14,17]. The economic model evaluating the profitability of the plant is based on equipment sizing and costing taking into account the operating conditions. The superstructure approach allows a flexible and systematic analysis of different process configurations. Sets of optimal design solutions are generated by the simultaneous optimization of the process in terms of thermodynamic performance and economic performance as a function of the decision variables. The multi-objective optimization is carried out by an evolutionary algorithm [18].

3. Process description

The overall process of thermo-chemical conversion of biomass into liquid fuels consists of: feed preparation, gasification, gas cleaning and treatment, and fuel synthesis and purification. Fig. 1 represents the unit operations of the process, as well as the energy integration options. It is focused on two process configurations shown in the superstructure; the first, employing a FICFB gasifier and the second, an EF gasifier. These configurations are referred to as FICFB and EF configuration.

3.1. Thermo-economic models

The thermo-economic models used in this study are based on the work of Tock et al. [13]. The models used for biomass pre-treatment, gasification and gas cleaning are identical to those previously developed by Gassner et al. [14,19].

3.1.1. Thermo-chemical conversion models

The biomass supplied to the process (50% humidity) is dried in an air drying unit which is optimized in respect to the residual humidity ϕ_d , wood and the inlet air temperature T_d . The biomass residual humidity at the gasifier inlet affects the heat required for its evaporation above the pinch, and therefore the amount of syngas needed for gasification. As a consequence, the heat available below the pinch is modified and therefore also the integration with the steam network. The dried biomass is then directly ground for FICFB gasification or torrefied ($T_{\text{t, out}} = 260$ °C) in order to be pulverized as required by the EF gasification. The model of the torrefaction unit is based on simple conversion ratios [20]. The FICFB gasifier consists of an indirectly heated circulating fluidized bed where the heat required for gasification is provided by circulating the bed material between two physically separated combustion and gasification chambers. The model of the FICFB gasifier is described in detail by Gassner et al. [19]. In the combustion

cleaning and conditioning. Their results show, for the present day design relying on an oxygen/steam blown circulating fluidized bed gasifier, an energy efficiencies of 47.8% and a chemical conversion efficiency of 50% (on an HHV basis and without considering the heat available for district heating). The associated production costs amount to 20 and 18 €₂₀₀₉/GJ_{LHV} (21 and 19 €₂₀₁₁/GJ_{LHV}) for the present day and for the near future design respectively [10]. These evaluations focus on case-studies operating at fixed conditions, for which only limited process integration has been taken into consideration. Recently Holmgren et al. [12] reviewed a series of studies about biomass gasification systems with methanol synthesis in terms of efficiency. The authors proposed several process configurations and on the basis of process integration evaluated the inclusion of different electricity production equipments and the effect of the moisture content of the raw material. Their base case configuration yields a chemical efficiency of 51% (on an LHV (lower heating value) basis). Comparing the results of the various studies on a common basis is a very difficult task, because of the different technologies considered, the different assumptions made and the degree of process integration. The objective of the present work is to systematically investigate the thermo-chemical conversion of lignocellulosic biomass into methanol applying multi-objective optimization techniques and including a detailed heat integration model to evaluate the potential for heat recovery and valorization. In the current study, the reference for heating values and efficiencies is on an LHV basis unless otherwise specified. The reference for currency is € in 2010, unless otherwise specified.

2. Methodology

The present work is based on a model superstructure that was previously developed to analyze and compare the thermo-chemical

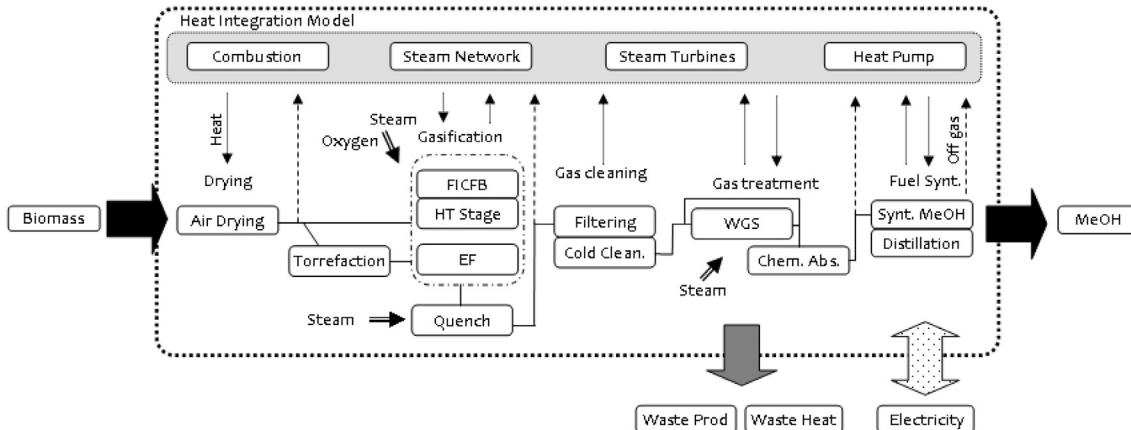


Fig. 1. Simplified process superstructure for MeOH production, including the energy integration options (full arrow: heat exchange, double arrow: steam and oxygen, broken arrow: possible streams that could be used as additional fuels). Waste heat from the plant may be coupled with a district heating system, but this option is not considered in the current study. The system boundary is represented by the external dotted line. Heat and mass balances within the system are always closed.

chamber ungasified char and fuels are oxidized with air to heat up the bed material which is transferred via a cyclone to a gasification chamber where steam reacts with the biomass feed to produce the syngas. The advantage of this gasification technology is that it produces an essentially nitrogen-free product gas without requiring air separation for the oxygen supply. The main disadvantages arise from the methane and tar content of the synthesized gas and from the high investments costs due to the complicated construction. A directly heated high temperature stage (HT stage) is introduced to reduce the methane and hydrocarbon content in the product gas through autothermal steam methane reforming. The heat for the endothermic reforming is thus satisfied by partial oxidation with pure oxygen. In the EF gasifier, the pulverized feed is entrained with the reacting gases, solid particles and gases move at approximately the same velocity. Consequently, smaller particles are required making the torrefaction step necessary. In this case, gasification is carried out using both oxygen and steam, and heat is provided directly by the oxidation of the feed. Advantages of this technology are the high capacity per unit volume (especially for the pressurized reactors) and the simpler geometry [21] (relatively to a fluidized bed). Because of the high temperature (1350 °C), the product gas is almost tar-free and a leach resistant molten slag is produced [22]. Disadvantages include the high oxygen consumption and a higher conversion of the energy of the feed into sensible heat [23]. The main operating conditions for the FICFB gasifier and the EF gasifier configurations are summarized in Table 1.

For both gasification configurations the product gas is quenched with steam to a temperature of 800 °C. In the gas cleaning step the product gas is cooled to 150 °C before entering the filter and the scrubber where it is cooled to ambient temperature. The WGS (water gas shift) reactor and the AGR (acid gas removal) step are used to bring the synthesis gas to the specifications required for the synthesis of methanol, that is to a stoichiometric ratio s ($s = (H_2 - CO_2)/(CO + CO_2)$) of 2. The exothermic WGS reaction produces extra H_2 (and CO_2) at the expense of CO by the addition of steam. It has been shown that, for kinetic reasons and in order to control by-products, a value slightly greater than two is preferred [5]. Lurgi reports a value between 2 and 2.1 [24]. The CO_2 concentration is typically adjusted to 4–8% for optimal activity and selectivity [25,5]. Furthermore, an excess of steam is required to allow an almost complete conversion of CO and to push the reaction away from solid carbon formation. The molar steam to carbon ratio is usually between 2 and 6, depending on the feedstock and reactor conditions [5]. In the model a steam to carbon (mainly) ratio of 2.5

was used. The absence of solid carbon at reactor conditions and at thermodynamic equilibrium is verified using the software Gemini [26], which calculates the equilibrium composition by minimization of the Gibbs energy of the system. The absence of solid carbon is verified at equilibrium but this does not guarantee that any carbon soot is produced. In order to maintain the steam to CO ratio and obtain the required gas compositions only part of the stream needs to be shifted. The WGS unit is modeled as a single reactor optimized with regard to the water gas shift reaction temperature T_{WGS} and the inlet temperature ($T_{in,WGS} = T_{WGS} - \Delta T_{WGS}$). The operating range between 250 and 320 °C is characteristic for intermediate temperature reactors [27]. The AGR step is modeled taking into consideration the values for the energy integration and economic analysis for chemical absorption as described by Tock [28]. Methanol synthesis is modeled by a multistage reactor with four beds in series [13,26,29] and it is optimized in respect to the synthesis gas inlet temperature $T_{m,in}$ and the reactors temperature and pressure

Table 1
Main operating conditions and decision variables with their variation range.

Section	Description	Variable	Unit	Value/range
Drying	Air inlet T	T_d	°C	[180 240]
	Wood ϕ at outlet	$\phi_{d, wood}$	%	[10 35]
Torrefaction	Torrefaction T	$T_{T, out}$	°C	260
Gasifier (FICFB)	Steam to biomass ratio	$R_{s/b}$	—	0.5
	Gasification P	p_g	bar	1.15
	Gasification T	T_g	°C	847
	HT stage	T_{HT}	°C	1350
	Steam preheating T	T_{steam}	°C	400
Gasifier (EF)	Steam to biomass ratio	$R_{s/b}$	—	0.6
	Gasification P	p_g	bar	30.15
	Gasification T	T_g	°C	1350
Water–gas shift	Delta inlet reactor T	ΔT_{WGS}	°C	[0.1 50]
	WGS reactor T	T_{WGS}	°C	[250 320]
	Steam to CO for WGS	$R_{s/CO}$	—	2.5
Methanol synthesis	$S = (H_2 - CO_2)/(CO + H_2)$	S	—	2.05
	Methanol synthesis inlet T	$T_{m, in}$	°C	[227 387]
	MeOH process P	p_m	bar	[75 90]
	MeOH synthesis T	T_m	°C	[252 267]
	Recycled fraction	R_m	mol	0.95
Steam network	Steam production P	p_{sp1}	bar	[40 120]
	Steam production P additional level	p_{sp2}	bar	[40 120]
	T of additional steam consumption level	T_{sc2}	°C	[50 250]
	Steam superheating T	T_{SH}	°C	[350 550]

(T_m, P_m). The operating range consider CO ed is close to the nominal conditions reported by Tock et al. [13] and within the characteristic operating conditions for conventional methanol production (50–100 bar, and 250–300 °C) [24,30]. A fraction of the off-gases ($R_m = 0.95$) is recycled into the synthesis reactors, to increase methanol conversion. In order to increase the purity of the produced methanol a final purification step is required. Two distillation columns allow achieving a methanol purity of over 99% (Tock [28]). Oxygen is required for the EF gasifier and for the directly heated high temperature stage of the FICFB gasifier. Oxygen for gasification is conventionally produced by pressure swing adsorption or cryogenic distillation. An ASU (Air Separation Unit) is not included in the current model superstructure but it will be integrated in future studies. The energetic and economic costs of the oxygen supply have been considered. From an economic standpoint oxygen is considered as a utility, purchased at the price indicated by Kirschner [21] as a function of the consumption rate. The range of values is reported in Table 2. The energetic cost of oxygen is taken into account considering an electricity consumption of 1080 kJ/kg_{O₂} as estimated by Hamelinck et al. [31] for cryogenic off-site oxygen production. Oxygen is delivered at standard ambient temperature and pressure. The decision variables relative to the integration of the steam network, for the heat integration model, include two steam production pressures (p_{sp1}, p_{sp2}) and one steam consumption temperature (T_{sc2}).

The reference scenario considered for the thermo-chemical conversion of biomass into methanol is a 20 MW_{th} sized plant. This capacity is very small for a plant producing bulk chemicals and may be considered representative of a demonstration plant. The main results are therefore also reported for a 200 MW_{th} plant. Nevertheless it should be considered that plant size may be limited, in this case, by biomass availability and logistics [32]. The main fixed operating conditions and the decision variables are summarized in Table 1. The ranges considered for the multi-objective optimization variables are based on literature and technological constraints.

3.1.2. Economic evaluation

The economic performance is evaluated by the total production cost including investment and operating costs. The capital cost estimates provide a basis for the overall comparison by assessing the trends implied by the decision variables, rather than an accurate estimate of the project. The cost estimation approach follows the one adopted by Gassner et al. [33] and Tock et al. [13] relying on data available in the literature. The currency exchange rates used are the yearly average exchange rates for 2010 [34] and all costs have been updated to year 2010 by using the Marshall and Swift

Index. The investment costs are calculated on the basis of the methodology outlined in Refs. [35,36]. The major process equipments are roughly sized and their purchase cost is calculated and adjusted to account for specific process pressures and materials using correlations from literature. The total investment cost is then calculated using multiplication factors to take into account indirect expenses like labor, transportation, fees, contingencies and auxiliary facilities. The operating costs [€/GJ_{MeOH}] take into account the cost of labor, maintenance (5% of the total investment), raw materials (biomass, oxygen) and utilities (electricity). The production cost [€/GJ_{MeOH}] is the sum of the operating cost and the depreciation cost, the latter being the total investment cost divided by the present worth of annuity (1) (depending on the investment rate ir and the economic lifetime t) and the yearly production of methanol. The interest rate ir and the economic lifetime t , were selected for continuity with previous studies [13,33], they correspond to a risk-free utility configuration, for example, representing the case of a government investment.

$$\text{Present worth of annuity} = \frac{(1+ir)^t - 1}{ir(1+ir)^t} \quad (1)$$

The main assumptions for the economic evaluation are summarized in Table 2.

3.2. Process performance indicators

To assess the process performance, thermodynamic, economic and environmental indicators can be defined. The considered indicators are:

Energy efficiency:

$$\eta_{en} = \frac{LHV_{MeOH} \cdot \dot{m}_{MeOH} + \dot{E}^-}{LHV_{Biomass,in} \cdot \dot{m}_{Biomass,in} + \dot{E}^+} \quad (2)$$

Chemical Efficiency:

$$\eta_{chem} = \frac{LHV_{MeOH} \cdot \dot{m}_{MeOH}}{LHV_{Biomass,in} \cdot \dot{m}_{Biomass,in}} \quad (3)$$

Equivalent efficiency:

$$\eta_{en_eq} = \frac{LHV_{MeOH} \cdot \dot{m}_{MeOH} + \frac{1}{\eta_{cc}} \Delta \dot{E}^-}{LHV_{Biomass,in} \cdot \dot{m}_{Biomass,in}} \quad (4)$$

where the superscripts – and + refer respectively to produced (output) and consumed (input) services, and all energy values are referred to an LHV basis. The equivalent energy conversion efficiency aims at correctly assessing the value of the produced or consumed by-products. In (4), contrary to definition (2), the consumed amount of power at the denominator is omitted and represented by the net overall output of electricity (\dot{E}^-) at the numerator [20]. The electrical power required is substituted by the equivalent amount of SNG (synthetic natural gas) which would be used for its generation in a CC (combined cycle). The economic indicators are the total investment and the production cost previously described in € referenced to the year 2010. The only environmental indicator taken into consideration is the yearly avoided CO₂ emissions [kt CO₂/year] obtained by the substitution of conventionally produced methanol with the biomass derived methanol. The account of the CO₂ emissions assumes that the combustion of the biomass and derived methanol is carbon neutral, while it takes into account the emissions due to the harvesting and transport of biomass and the consumption of electricity which is dependent on the electricity mix. The CO₂ emissions relative to the

Table 2
Economic evaluation assumptions.

Parameter	Value
Marshall and Swift index (2010)	1473
Dollar exchange rate (€_US\$)	1.5 US\$/€
Expected lifetime	15 years
Interest rate	6%
Plant availability	90%
Operators ^a	4 p./shift
Operator's salary	66000 €/year
Wood costs ($\phi_{wood} = 50\%$)	6€/GJ _{BM} (21€/MWh _{BM})
Electricity price ^b	43 €/GJ (155€/MWh)
O ₂ costs (1–10 ⁵ m ³ /h) [21]	0.03–0.7 €/kg

^a For a plant size of 20 MW_{th}, biomass input. For other production scales, an exponent of 0.7 with respect to plant capacity is used. The prices of electricity and biomass are representative of the European market.

^b The high price of electricity accounts for the production/consumption of 'green' electricity.

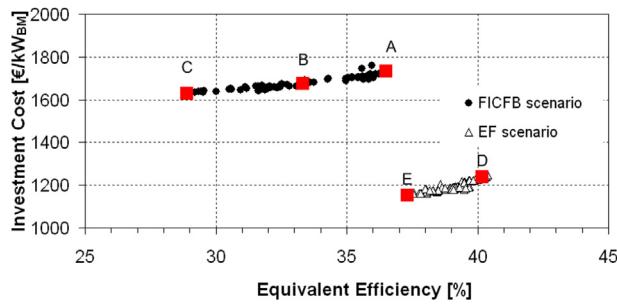


Fig. 2. Optimization results for the FICFB and the EF configurations.

use of fossil derived methanol take into account its production and combustion. The data used for environmental evaluation refers to the Swiss context and are taken from Ref. [37].

4. Optimization

The multi-objective optimization was carried out selecting as the objective functions, the minimization of the capital investment cost and the maximization of the equivalent efficiency (4). This is done in order to generate a list of optimal processes among which the best solution (i.e. the one that minimizes the total production cost) may be selected for given economic conditions (i.e. cost of resources, electricity price, interest rate, expected lifetime etc.). The generated Pareto fronts for the two conversion configurations employing the EF and the FICFB gasifiers are represented in Fig. 2, in terms of specific investment cost (€/kW_{BM}) and equivalent efficiency. They represent the family of optimal solutions for which efficiency can't be improved without a raise of the investment cost, and vice versa investment costs can't be lowered without a loss in efficiency. The reference plant capacity in Fig. 2, is 20 MW_{th}. Results are summarized in Table 3.

The designs A, B, and C are shown as representative of a high efficiency – investment cost, intermediate efficiency – investment and low efficiency – investment cost optimal designs for the FICFB configuration. Designs D and E are representative of a high efficiency – investment cost and low efficiency – investment cost optimal designs for the EF configuration. The values of the operating conditions for these configurations and the distribution of the optimization variables for all configurations obtained in the Pareto front are reported in the Supplementary Material section.

4.1. Analysis of the optimization results

The results show that the energy efficiency of the configurations presented is very similar, while the equivalent efficiency highlights

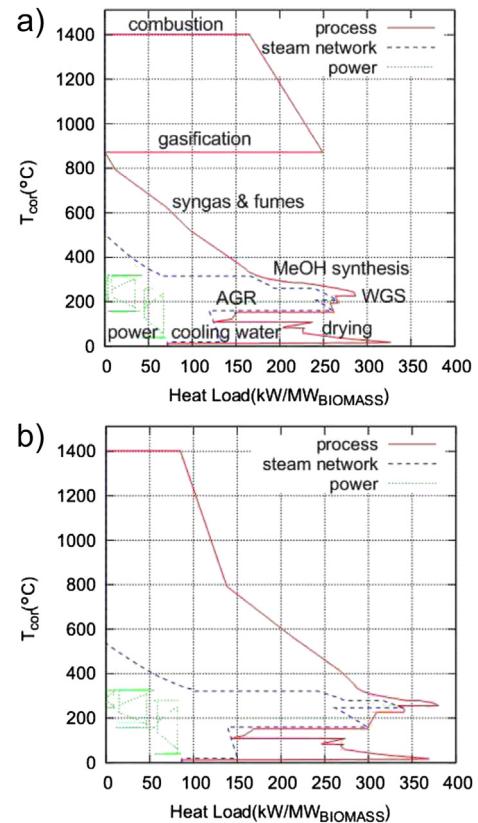


Fig. 3. Comparison of the composite curves of (a) design A, with a FICFB gasifier, and (b) design D with an EF gasifier.

the different capacity for co-production of electricity. The total production costs also result similar within the solutions for each configuration, this is a consequence of the small spread obtained for the investment costs for both configurations (about 100 €/kW for both cases at 20 MW_{th} capacity) but also the trade-off between increasing investment costs and decreasing operating costs with improving efficiency. Production costs are lower for the EF configuration, and the difference becomes even more substantial when considering a higher capacity plant. This is because EF gasifiers may be built in larger units, while several gasification units are required for the more complicated FICFB design in order to attain larger capacities. The comparison of the composite curves of designs A and D is represented in Fig. 3. Design A, with a FICFB, presents a pinch at the gasification temperature, while in design D the pinch disappears because for the EF gasifier the heat is directly provided by the partial oxidation of the feed.

Table 3
Summary of results for different design solutions.

Design	Cap. ^a MW _{th,BM}	η_{en_eq} %	η_{en} %	η_{chem} %	Steam cycle power kW/MW _{BM}	Elec net cons ^b kW/MW _{BM}	Inv cost €/kW _{BM}	Op costs €/GJ _{MeOH}	Prod costs €/GJ _{MeOH}
<i>FICFB</i>									
A	20/200	36.50	42.55	44.71	70.3	70.3(15.6)	1736/1322	29.6/24.78	44.6/36.6
B	20	33.31	44.78	49.18	37.2	98.4	1677	30.9	44.0
C	20/200	28.88	44.62	50.64	7.5	134.9	1632/1282	32.9/28.9	45.4/38.8
<i>EF</i>									
D	20/200	40.15	44.28	45.84	84.8	35.3 (38.5)	1241/683	24.1/18.7	34.6/24.5
E	20	37.30	43.12	45.25	68.7	49.2	1155	25.4	35.3

^a For designs A, C and D results are reported for plant capacities of 20 MW_{th} and 200 MW_{th}.

^b The share of power required for oxygen production is reported in parenthesis. Design D results in a net exporter of electricity, if the electricity required for oxygen production is not accounted for. The electricity consumption due to oxygen production is similar across the FICFB and the EF configuration. The oxygen requirement for a capacity of 20 MW_{th} is of 0.28 kg/s and 0.47 kg/s for the FICFB and the EF configurations respectively.

The analysis of the optimization results of both configurations suggests that the co-generation of electricity has a determining importance in the positioning of the designs on the Pareto front for both gasifier options. Designs with the lowest efficiency are the ones with smaller units for heat recovery and electricity generation, which are also the cheapest ones. Moving from the lowest efficiency designs to the highest ones, the amount of electricity produced increases, the process remaining a net importer of electricity all along the Pareto front. This can be visualized by the composite curves of the three designs A, B, and C represented in Fig. 4. The role of the energy integration is also shown in Fig. 5 where electric power and total electricity required by the process are represented as a function of the equivalent efficiency (4) for the FICFB and the EF configuration. The secondary axis indicates the chemical (2) and energy (3) efficiencies. These results show the importance of the heat integration and the cogeneration of electricity in improving

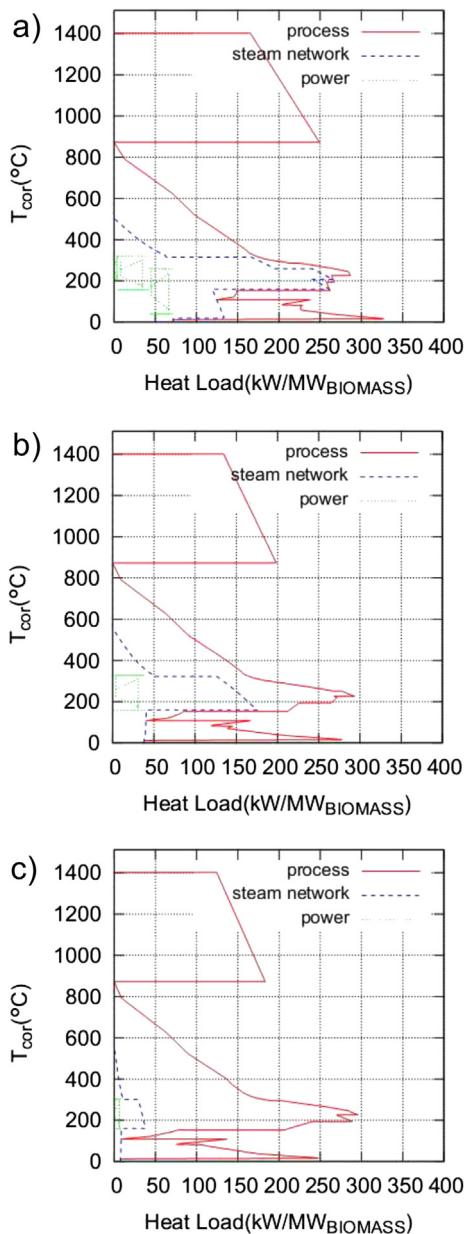


Fig. 4. Composite curves of designs A (a) B (b) and C (c) belonging to the FICFB configuration.

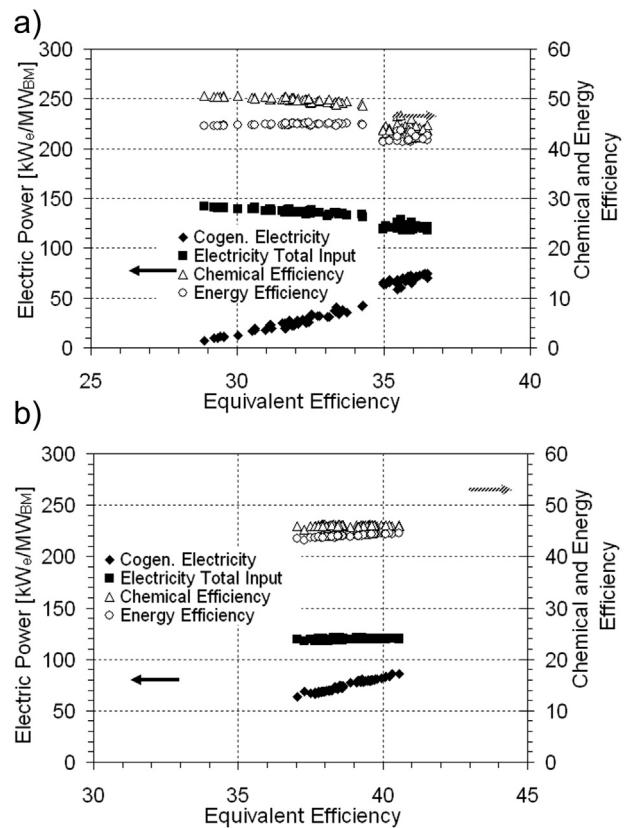


Fig. 5. Effect of heat integration on the optimal conceptual designs for the FICFB configuration (a) and the EF configuration (b). The cogeneration power, total electricity requirement, the chemical and energy efficiency are represented as a function of the equivalent efficiencies. The x-axes do not have the same scale.

the overall efficiency of the process. For both design solutions the highest equivalent efficiency processes are the ones displaying a higher fraction of the required electricity produced via a Rankine cycle. The overall equivalent energy efficiency results are higher for the EF designs than for the FICFB. This is in part due to the integration of the combined cycle which is able to provide a larger fraction of the electricity requirement. The EF gasifier, in fact, makes available a higher fraction of the feed as high temperature heat which may be converted into electricity. As explained before, an inconvenient of the FICFB gasifier is the presence of methane and tars in the produced gas. In this study tar removal and methane reforming is carried out by an HT stage following the FICFB gasifier. The temperature of the HT stage depends on technological constraints and the nature of the biomass resource. Its value greatly affects the performance of the process, as higher temperatures impose larger oxygen consumption (and therefore energy requirements). For design A, for example, the equivalent efficiency $\eta_{en, eq}$ could be raised by 1.7% point to 38.2% ($\eta_{en} = 44.2\%$, $\eta_{chem} = 46.5\%$) if the HT stage temperature was reduced by 100 °C–1250 °C. In a subsequent optimization study the influence of the temperature can be studied in more detail. Other technological options for the reduction of tars and the reforming of methane have been investigated in other studies, such as catalytic cracking and mechanism methods (i.e. scrubbers). A review of tar reduction and control technologies for biomass gasification is presented by Han et al. [38]. The energy efficiencies are similar for both configurations and range between 42 and 45%. The chemical efficiencies range between 45 and 51%, corresponding to mass yields between 42 and 48%. These values are in the same range of the efficiencies

reported in the literature but the comparison results difficult because of the different process options and operating conditions. The efficiency of the FICFB is slightly lower than what is reported by Hamelinck et al. [7] for a fluidized bed, this difference is mainly due to the presence of the HT stage in the present study.

The investment cost build-up for the high efficiency designs of the FICFB configuration and the EF configuration are shown in Fig. 6. The cost of the gasifier represents a large fraction of the total investment cost. The higher cost for the FICFB is in large part due to the cost of the gasifier itself. The lower estimated cost of the EF gasifier is due to its simpler design and the possibility of building larger units. For the FICFB configuration, in fact, two gasification units are required, while only one gasifier is required in the EF configuration (for the 20 MW_{th} capacity). The production costs results are lower for the EF gasifier mainly because of the impact of the lower investment costs. The oxygen required by the processes appears in the production costs as it is purchased [21]. This is why design D results a slight net exporter of electricity. Nevertheless, for a more reliable comparison of the cost of the two technologies more information would be needed.

5. Sensitivity analysis

The base price considered for biomass is 6 €/GJ and for electricity, 43 €/GJ Table 2. For comparison, the price of electricity for industry in France is about 20 €/GJ [39], but in Italy and Switzerland about 40 €/GJ [39,40]. Furthermore, if the product is to be considered renewable, the imported electricity should also be provided by a renewable resource, which may result in a higher electricity price. The price considered for biomass is also variable across the literature. Huisman et al. [10] consider 20 €/MWh (about 6 €/GJ), Tock et al. 33 €₂₀₀₉/MWh (about 10 €/GJ). The sensitivity analysis was carried out considering a price range for electricity between 20 and 50 €/GJ and for biomass between 3 and 12 €/GJ. The sensitivity of the production and operating costs of the electricity and biomass prices for the three previously described designs, belonging to the FICFB configuration is represented in Fig. 7. The strong impact of the biomass cost highlights the importance of the chemical efficiency to obtain favorable methanol production costs.

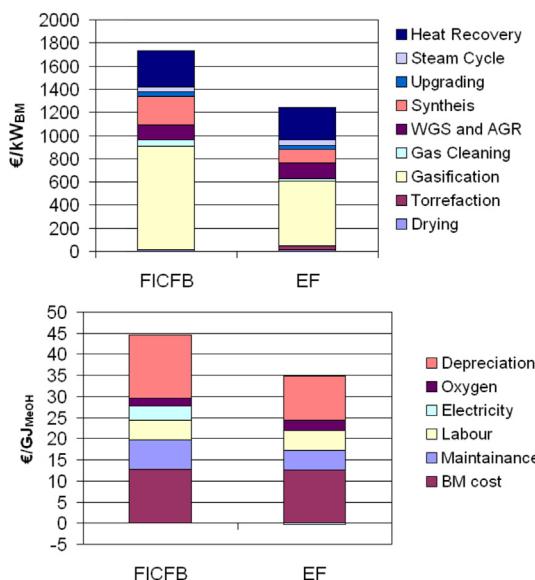


Fig. 6. Investment and production cost build-up for the FICFB (design A) and the EF (design D) configurations, 20 MW_{th} capacity.

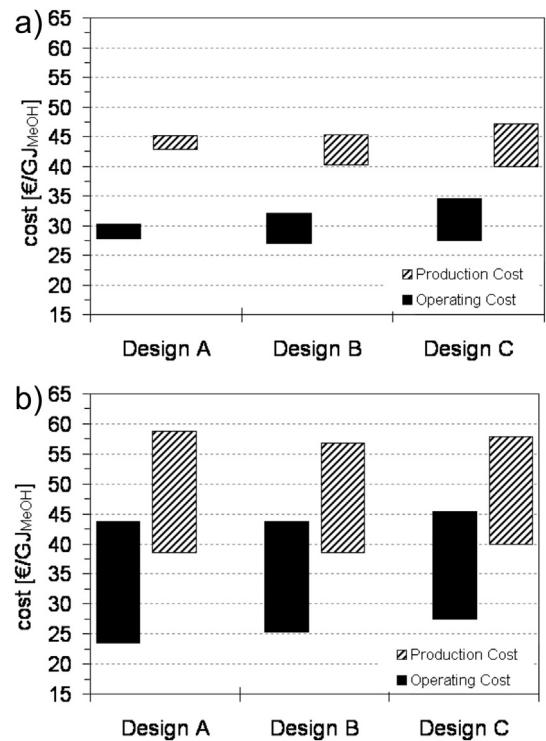


Fig. 7. Sensitivity analysis for the FICFB configuration a) base price for biomass is 6 €/GJ, electricity price range 20–50 €/GJ. b) base price for electricity is 43 €/GJ, biomass price range 3–12 €/GJ, 20 MW_{th} capacity.

The reference configuration considered for this study is a 200 MW_{th} sized plant. Fig. 8 shows the variation of operating and production costs and the environmental impact in terms of yearly CO₂ avoided emissions as a function of the plant size, for designs A

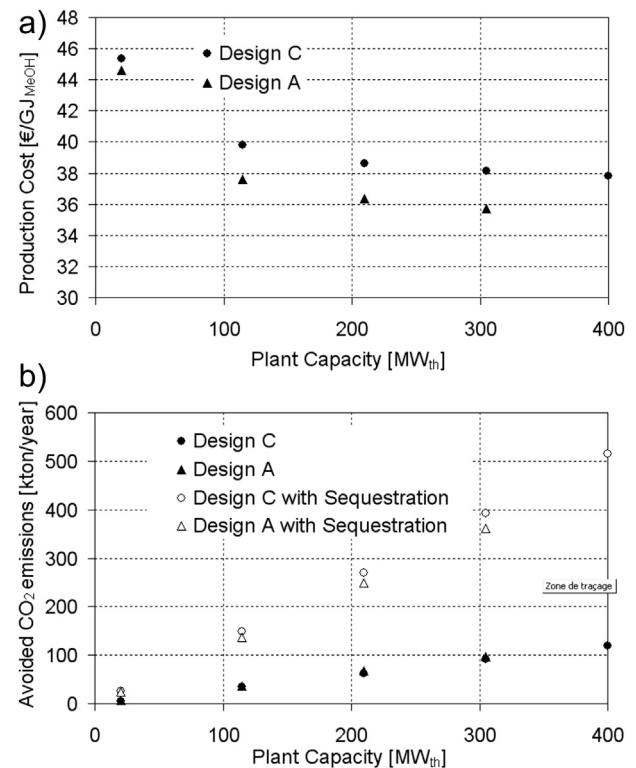


Fig. 8. a) Production cost and b) avoided CO₂ emissions – sensitivity analysis on the plant capacity.

and C. As expected, the economies of scale have non-linear impact on the production costs which decrease by about 15% from a plant capacity of 20 MW_{th} of biomass input to about 200 MW_{th}. The production cost reduction between 200 MW_{th} and 400 MW_{th} is only about 2%. The avoided CO₂ emissions increase linearly with the plant capacity as they are proportional to the produced methanol. The distance at which biomass is sustainably available also affects the evaluation of the production cost and the CO₂ emissions in terms of the optimal plant capacity, but it hasn't been the focus of this study.

6. Conclusions

The thermo-economic optimization of biomass thermo-chemical conversion into methanol was carried out considering as alternative configurations, a FICFB gasifier and an EF gasifier. Multi-objective optimization and process integration allowed to consistently compare the performances of the optimized conceptual designs in terms of efficiency and costs. The results show the importance of the energy integration and in particular of the Rankine/cogeneration cycle for the improvement of the overall efficiency of the process. The EF configuration displays higher equivalent efficiencies in comparison to the FICFB configuration because the integration of a steam cycle allows for a larger production of electricity, satisfying part of the energy requirement of the plant. On the other hand, the overall energy efficiencies are similar for the two configurations and result of about 43–45%. The chemical efficiencies range between 45 and 51%. From an economic standpoint, the production costs range from about 35 to 45 €/GJ for the 20 MW_{th} case and from 25 to 35 €/GJ for the 200 MW_{th}. Production costs remain well above the current price for natural gas derived methanol (16 €/GJ [2]), and are strongly influenced by the estimate of the cost of the gasification unit and biomass purchase price. Nevertheless the evaluation of the avoided CO₂ emissions highlights the potential of this biomass conversion route in a carbon constrained world.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.energy.2013.05.029>.

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