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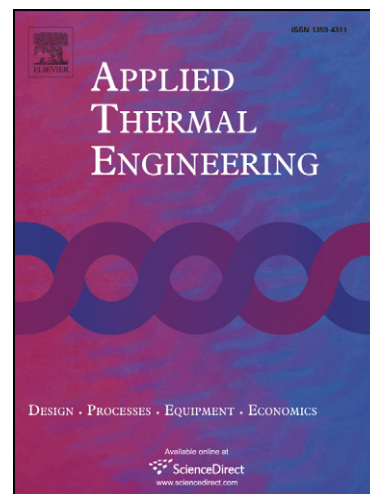
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**Application of exergy balances for evaluation of process configurations
for biological hydrogen production**

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Abstract

Exergy analysis was applied to a novel process for biological production of hydrogen from biomass employing thermophilic and photo-heterotrophic bacteria. The exergy content of the process streams is calculated using a MS-Excel spreadsheet.

The scrutinized process incurs an exergy loss of 7-9 % of the total exergy input. The efficiency based on chemical exergy of biomass feed and produced pure hydrogen refers to 36-45% depending on the configuration of the overall process. The results presented in the paper underline the strong dependence of obtained exergetic efficiency from definition of products and shows options for process improvement and optimization.

Keywords: biohydrogen, process integration, exergy analysis, exergetic efficiency, process simulation, renewable energy, biofuel

1. Introduction

Hydrogen will be an important energy carrier in the future. Presently hydrogen is almost completely produced from fossil fuels or by electrolysis of water. To make the future hydrogen economy fully sustainable, renewable resources instead of fossil fuels have to be employed for hydrogen production. Besides biomass gasification, hydrogen from biomass can also be produced in a non-thermal way using bacteria.

During the last years, different anaerobic and photo-heterotrophic fermentation processes were investigated to produce biohydrogen. However, single stage processes currently do not work economically. A promising way for the production of hydrogen from biomass in a non-thermal way is a 2-stage bioprocess consisting of a thermophilic fermentation step followed by a photo-heterotrophic fermentation [1-4].

Most research currently is performed concerning the selection of microorganisms, optimization of yield and rate of hydrogen production as well as reactor design. Only a few studies are aiming at the design of the whole production process including feedstock pre-treatment and gas-upgrading as well as additional process steps to successfully combine both fermentation processes and remove hydrogen from the fermentation broth.

To obtain an economic and competitive overall process for the biological production of hydrogen from biomass, careful selection of upstream and downstream processes as well as optimal integration of all steps in terms of minimizing residual streams and heat demand is crucial. Besides mass and energy balance, exergy analysis will be applied to

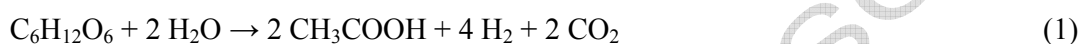
the novel process for non-thermal production of hydrogen from biomass as a selection criterion for the most efficient hydrogen production route.

2. Hyvolution Process

The novel approach for the non-thermal hydrogen production from biomass in HYVOLUTION is based on a combined bioprocess employing thermophilic and photo-heterotrophic bacteria, to provide a high hydrogen production efficiency (see Fig. 1) [5].

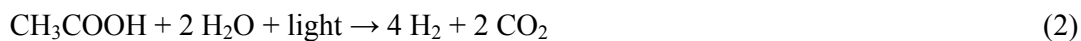
The process starts with the necessary pre-treatment of biomass to provide a suitable feedstock for thermophilic fermentation (THF). Starch and sugar containing as well as lignocellulosic biomass will be considered as feedstock. The ultimate objective finally is the use of residues from agriculture and food processing (molasses, potato steam peelings, wheat bran, straw) for the production of biohydrogen. For a first evaluation of the overall process, a starch-based feedstock was selected, represented by wheat, due to the known pre-treatment procedure. For feedstock wheat, a dry matter content of 90% as well as a starch content of 70% based on dry matter was assumed. The pre-treatment of wheat is a conventional and proven liquefaction and saccharification process. The milled feedstock is first mixed with water to 35 wt% solid mixture. α -amylase is added and the mixture is heated to 105°C with direct steam and kept at 95°C for 2 hours. Finally, the liquefied feedstock is fed to the saccharification reactor and mixed with gluco-amylase. A residence time of 72 hours at 60°C gives an overall conversion of starch in the pre-treatment of about 97%.

In the first fermentation step, thermophilic bacteria growing at temperatures of at least 70°C produce hydrogen gas and organic acids as the main by-products. Depending on the fermentation pathway of the bacteria and built by-products, different amounts of hydrogen per mole of sugar are yielded. Assuming that glucose is the substrate and acetic acid is the main by-product, the thermophilic fermentation can be represented by the following reaction (Eq. (1)):



Experimental results in the thermophilic fermentation step requested a decrease of hydrogen partial pressure in the fermenter due to inhibitory effects. In case of glucose-based feedstock, a maximum hydrogen partial pressure of 20 kPa was identified giving optimum productivities and yields under the applied conditions [3, 6]. To lower the hydrogen partial pressure in the thermophilic fermenter, it was suggested to apply gas-stripping. Since used thermophilic bacteria are very sensitive to oxygen, an inert gas has to be used. Nitrogen is not applicable since it can be hardly separated from hydrogen during gas-upgrading. Finally stripping with CO₂ was identified as a feasible option, since it is produced in the process and available after gas-upgrading.

Produced acetic acid (HAc) can be used as substrate for hydrogen production in a consecutive photo-heterotrophic fermentation step (PHF). Based on acetic acid as substrate the reaction (Eq.(2)) can be written as:



Through the combination of thermophilic fermentation with photo-heterotrophic fermentation, almost complete conversion of the substrate to hydrogen and carbon dioxide can be obtained, resulting in up to 75% conversion efficiency or 9 moles of hydrogen per mole of glucose [5, 7]. Basic process data for the thermophilic and photo-heterotrophic fermentation step used for calculation of mass and heat balances are summarized in Table 1.

To provide pure hydrogen, finally carbon dioxide has to be separated from produced gas. Due to fluctuations in quantity and quality of the raw gas produced in the bioreactors, a specific gas treatment is required. In this paper, the industrial state-of-the-art systems adsorption (VSA, vacuum swing adsorption) and amine absorption/desorption (MEA) were chosen for gas-upgrading since they are well-documented processes with well-known performance. Finally, a novel membrane contactor will be evaluated and integrated in HYVOLUTION process. In lab scale experiments, this system demonstrated highly efficient carbon dioxide separation from gas mixtures of biological origin [8].

Investigation and optimization of the different process steps within the HYVOLUTION project is mainly based on experimental work. Focus is given to the selection of microorganisms and the optimization of process parameters of the different process steps to increase hydrogen yield and productivity as well as to decrease by-product formation. However, optimization of single steps might not give a satisfactory overall process. Therefore, process simulation is used to combine and integrate the single

process steps and finally select process routes by comparing the performance of different unit operations and design options. The commercial software package ASPENplus® was selected to predict the behaviour of Hyvolution process using basic mass balances, energy balances, phase and chemical equilibrium as well as reaction kinetics. Software package and simulation models, which are based on experimental data, are described in [9, 10]. Mass and energy balances based on different substrate concentration in thermophilic and photo-heterotrophic fermenter are presented in [11] for starch based as well as lignocellulosic feedstock.

3. Exergy Analysis

In general, exergy analysis is applied to identify the type, location and magnitude of thermal losses [12]. Exergy is calculated as the sum of three components – chemical exergy, physical exergy and the exergy change of mixing. The total exergy flow rate of a material stream at actual conditions can be obtained from Eq. (3):

$$EX = F \cdot (Ex_{chem} + Ex_{phys} + \Delta Ex_{mix}) \quad (3)$$

Chemical exergy, physical exergy as well as exergy change of mixing are calculated following the procedure described in [13].

Chemical exergy of biomass (see Eq. (4)) can be estimated using lower heating values and data from elemental analysis [14, 15]:

$$Ex_{chem,bio} = x_{bio} \cdot (\beta \cdot LHV_{bio}) + x_S \cdot (Ex_{chem,S} - C_S) + x_{water} \cdot Ex_{chem,water} + x_{ash} \cdot Ex_{chem,ash} \quad (4)$$

The factor β is the ratio of the chemical exergy to the lower heating value (LHV) of the organic fraction of biomass and C_S is the calorific value of sulphur. Higher heating values (HHV) of biomass can be accurately calculated by the correlation developed by Channiwala and Parikh [16]. For further details on calculation of chemical exergy of biomass see [13, 17]

For real processes, the exergy input always exceeds the exergy output. This unbalance is caused by exergy losses due to irreversibilities (Eq. (5)):

$$\sum_{in} Ex_j + \sum_{in} (Ex_Q + Ex_W) = \sum_{out} Ex_k + \sum_{out} Ex_Q + I \quad (5)$$

Eq. (5) considers the exergy of all entering and leaving material streams, the sum of all thermal exergy and work interactions (Ex_Q and Ex_W) involved in a process as well as the irreversibility I of the system [18]. The exergy output usually consists of the exergy of product and waste streams leaving the system. Cornelissen [19] discusses three types of exergetic efficiency given in Eqs. (6a)-(6c). Simple exergetic efficiency expresses the ratio of exergy output (exergy of output streams) and exergy input (exergy of input streams). Eq. (6b) represents the exergy losses of the process. Rational exergetic efficiency (see Eq. (6c)) is initially defined by Kotas [20]. This efficiency is given by the ratio of exergy of product streams to the exergy input. Another possibility is to use the chemical exergetic efficiency, defined as the ratio between chemical exergy of product and chemical exergy of feedstock, presented in Eq. (6d).

$$\eta_{ex,1} = \frac{Ex_{out}}{Ex_{in}} \quad (6a)$$

$$\eta_{ex,2} = \frac{I}{Ex_{in}} \quad (6b)$$

$$\eta_{ex,3} = \frac{Ex_{prod}}{Ex_{in}} \quad (6c)$$

$$\eta_{ex,4} = \frac{Ex_{chem,prod}}{Ex_{chem,feed}} \quad (6d)$$

4. Mass and Energy Balances

Presented overall process balances are calculated for the production of 60 kg/h of pure hydrogen (99,7 vol%) corresponding to approx. 2 MW of thermal power (based on lower heating value) without considering heat integration and recirculation of effluents within the process. The overall balances include the increased feedstock demand when applying VSA for gas upgrading due to hydrogen losses in the VSA-unit (25%). The following process options are investigated:

- Feedstock wheat, no stripping, MEA
- Feedstock wheat, CO₂-stripping, MEA
- Feedstock wheat, no stripping, VSA

Key data concerning overall mass and energy balances for selected process options are summarized in Table 2.

Besides biomass feed and water demand, cumulative heat demand and minimum heat demand as well as estimates for the demand of electric power are given. Minimum heat demand follows from the construction of hot and cold composite curve (Q-T-graph).

“Cell Mass” and “Non-Fermentable” refers to biomass produced in the process in form of cell mass and remaining non-fermentable solids from feedstock, respectively.

Production of heat and energy from residues, by-products as well hydrogen losses are not considered yet.

For further details on heat and mass balances see [11, 21]

5. Exergy Balances and Exergetic Efficiency

A MS-Excel spreadsheet has been developed to calculate in a fast and systematic way the exergy of compounds and streams of Hyvolution process. The calculation of necessary thermodynamic properties are based on integrated polynomial functions for the values of specific heat, entropy and enthalpy, using the same correlations as in the used process simulation tool, to ensure full compatibility with the solutions for mass and energy balance. For more details on exergy calculation see [13, 17].

Fig. 2 shows the exergy balance of the process option without stripping and gas-upgrading by VSA obtained from the developed MS-Excel spreadsheet. It is important to consider the magnitude of exergy of the material streams involved in the process compared to the exergy of incoming and outgoing heat and work streams, resulting from the dominance of chemical exergy in the process.

5.1 Exergy of Overall Process

Fig. 3 shows the exergetic efficiency of the different process steps in terms of simple exergetic efficiency ($\eta_{ex,1}$), exergy losses also referred as irreversibilities ($\eta_{ex,2}$), rational exergetic efficiency ($\eta_{ex,3}$) and chemical exergetic efficiency ($\eta_{ex,4}$) according to Eqs.

(6a)-(6d). Products in the calculation of rational exergetic efficiency as well as chemical exergetic efficiency include the purified hydrogen stream and pure hydrogen, respectively. In case of rational exergetic efficiency the term product comprises the exergy of the whole product stream (stream “HYDROGEN”, see Fig. 2). This includes chemical exergy of hydrogen as well as remaining traces of impurities (CO₂ and water vapour), physical exergy and exergy change of mixing. In case of chemical exergetic efficiency, only chemical exergy of pure hydrogen is considered as usable product. A hydrogen content of 99.7 vol% is assumed in the product stream.

The results show that the highest efficiency is achieved for the process option without stripping and applying gas-upgrading by MEA-absorption. Largest exergy losses occur in the process option applying CO₂-stripping, but are comparable to the option without stripping and gas-upgrading by VSA. Since exergy losses for the option with CO₂-stripping are higher compared to cases without stripping, alternative measures for the reduction of hydrogen partial pressure in the thermophilic fermentation step are recommended from the exergetic point of view.

Compared to the process options with MEA-absorption, rational and chemical exergetic efficiency are considerably lower for cases applying VSA. Main reasons are the high hydrogen losses during regeneration of VSA and the connected higher feedstock demand to obtain 60 kg/h pure hydrogen. Results indicate that this process step needs improvement. Losses might be reduced by re-designing the process step and using inert gas instead of purified hydrogen for flushing during the regeneration of the adsorber. This will reduce, but not completely avoid hydrogen losses in this process step.

Exergy losses obtained for the investigated process options are between 7 % and 9 %.

The efficiency based on chemical exergy of biomass feed (wheat) and produced pure hydrogen refers to 36-45% depending on the configuration of the overall process. The obtained results correspond with data [18] for anaerobic digestion of biomass to H₂ as well as to biogas production with 36 % and 46%, respectively.

Exergetic efficiencies of further process options as well as of the different involved process steps are available from [17, 21]. Differences in results compared to the actual paper are due to different definition of especially rational exergetic efficiency as well as the fact of slightly changed stream composition for the removal of cell mass from the process streams.

5.2 Parameter Study on Product Definition

As seen in earlier work [11], obtained exergetic efficiency strongly depends on the definition of products. Figs. 4 and 5 present the results of a parameter study investigating the influence of definition of products on rational exergetic efficiency ($\eta_{ex,3}$) and chemical exergetic efficiency ($\eta_{ex,4}$), respectively. Results are used to find options for process improvement.

Case “H₂” describes the base case considering only pure hydrogen stream or pure hydrogen as product of the process as described above. Case “H₂+H₂(Tail)” describes an option where hydrogen in the tail gas of VSA step is used for example to produce heat and power by combustion. This case is only considered for chemical exergetic

efficiency ($\eta_{ex,4}$), since rational exergetic efficiency always considers the whole stream, including also the content of CO_2 (Case “ $H_2+H_2(Tail)+CO_2$ ”). Considering the tail gas as a product increases rational exergetic efficiency of the process option applying VSA from 25 % to 32 % due to the high hydrogen content of the tail gas, while the slightly increase of efficiency of the process options with MEA-absorption is caused only by CO_2 . Attention has to be given to the interpretation of the high chemical exergetic efficiency ($\eta_{ex,4}$) of the process options applying CO_2 -stripping in Fig. 4. The high efficiency for cases considering CO_2 as a usable product only results from definition of $\eta_{ex,4}$, where all CO_2 leaving the process is considered as a product, but entering stripping gas is not included in the input term. As defined above (see Eq. (6d)), only chemical exergy of used feedstock is considered in the input term for calculation of this efficiency.

A significant increase of exergetic efficiency of the overall process can be achieved, when defining remaining biomass as usable product of the process. Exergetic efficiencies almost double as follows from Figs. 4 and 5. Remaining biomass refers to biomass produced in the process in form of cell mass and non-fermentable solids from feedstock, respectively. Cell mass produced during fermentation might be re-introduced to the process as feedstock or source of nutrients or might be used to produce heat and power together with the non-fermentable fraction. A separate investigation was not possible, since so far a separation of non-fermentables after pre-treatment step is not foreseen in the process. Non-fermentables are removed after thermophilic fermentation together with the cell mass produced in this process step.

Introducing released heat to the portfolio of usable products shows that from the exergetic point of view the contribution of heat integration to the increase of exergetic efficiency of the process is negligible due to the strong impact of chemical exergy compared to physical exergy. Nevertheless, heat integration plays an important role from the point of view of energy demand and economic evaluation (see Table 2 as well as [11]).

A further increase of exergetic efficiency will result from the (re-) use of process effluents to replace tap water or the application of effluents as a liquid fertilizer in agriculture.

6. Conclusions

Exergy analysis was applied to a novel process for biological production of hydrogen. The exergy content of the process streams was calculated using a MS-Excel spreadsheet showing good agreement with literature.

The process scrutinized by the Hyvolution project incurs an exergy loss of 7-9 % in reference to the total exergy input. A chemical exergetic efficiency of 36-45% was obtained without considering any heat and process integration.

A parameter study underlines the strong dependence of obtained exergetic efficiency from definition of obtained products and shows options for process improvement and optimization. Following the results, it is recommended to avoid hydrogen losses or to use the chemical exergy of hydrogen in the tail gas to produce heat and power for the process. Most important contribution to an increase of exergetic efficiency comes from

(re-) use of produced cell mass and non-fermentables as well as effluent from process as feedstock or nutrient, for heat and power generation or fertilizer, respectively. From the exergetic point of view, the contribution of heat integration to the increase of exergetic efficiency is negligible for the investigated low temperature process.

The calculated improvement of exergetic efficiencies only represents a theoretical maximum. Impact on exergy balance and exergetic efficiency has to be investigated in more detail considering also additional process steps necessary to implement the suggested process improvements.

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Nomenclature and Abbreviations

C Calorific value, kJ/kg

Ex Molar exergy, kJ/mol

EX Exergy flow rate, kJ/s

F Molar flow rate, mol/s

I Irreversibility

HAc Acetic acid

HHV Higher heating value of biomass, kJ/kg

LHV Lower heating value of biomass, kJ/kg

MEA MEA (Mono-Ethanol-Amine)-absorption/desorption

PHF Photo-heterotrophic fermentation

PRE Pre-treatment

THF Thermophilic fermentation

VSA Vacuum swing adsorption

x Mole fraction

β Ratio of the chemical exergy to LHV

η Efficiency, Loss

Subscripts

ash Ash

bio Biomass

chem Chemical

ex Exergetic

feed Feedstock

in Input

mix Mixing

out Output

phys Physical

prod Product

Q Heat

S Sulphur

water Water

W Work

Fig. 1. Scheme of Hyvolution-process.

Fig. 2. Exergy balance of process option without stripping; gas-upgrading by VSA [MJ/h].

Fig. 3. Overall exergetic efficiency of investigated process options.

Fig. 4. Parameter study on product definition – Rational exergetic efficiency (Efficiency 3).

Fig. 5. Parameter study on product definition – Chemical exergetic efficiency (Efficiency 4).

Table 1

Basic settings for pre-treatment, thermophilic fermentation (THF) and photo-heterotrophic fermentation (PHF) for feedstock wheat.

Plant capacity	60 kg/h hydrogen
Feedstock	wheat
Conversion starch	97 % (wt)
Conversion substrate to hydrogen	80 % (wt) per fermentation step
Conversion substrate to cell mass	15 % (wt) per fermentation step
Substrate losses	5 % (wt) per fermentation step
Temperature THF	70 °C
pH THF	6.5
Substrate concentration THF	50 g/l glucose
Temperature PHF	30 °C
pH PHF	7.3
Substrate concentration PHF	100 mM acetic acid

Table 2

Key data concerning overall mass- and energy-balances for selected process options.

		No stripping, VSA	No stripping, MEA	CO ₂ -stripping, MEA
Flow rate feedstock	[kg/h]	1212	970	970
Flow rate water	[kg/h]	70000	56000	56000
Flow rate strip-gas (CO ₂)	[kg/h]	0	0	1710
Flow rate cell mass	[kg/h]	103	82.5	82.5
Flow rate non-ferment.	[kg/h]	295	236	236
Concentration H ₂ raw gas	[mol/mol]	64.0	64.0	34.0
Electric power	[kW]	210	70	120
Heat flow	[kJ/s]	1430	1650	2570
Min. heat flow	[kJ/s]	1230	1100	2200
H ₂ losses	[%]	25	0	0

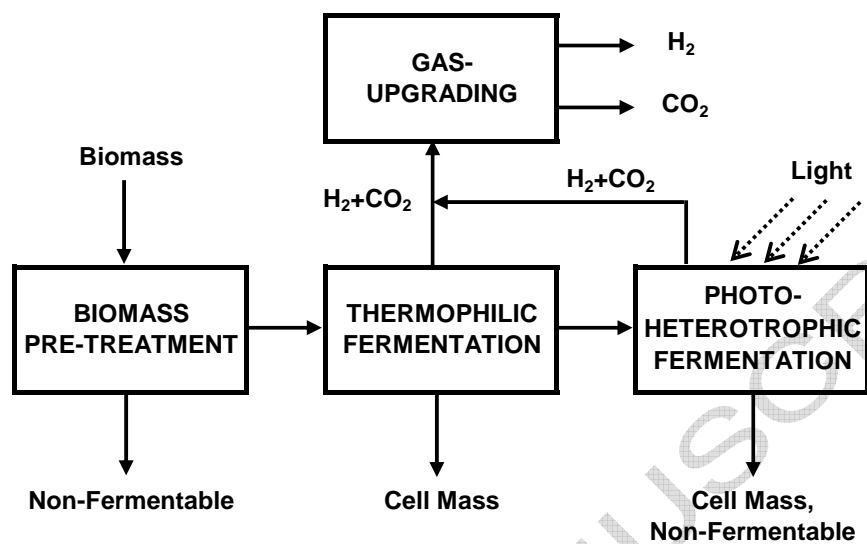


Fig. 1. Scheme of Hyvolution process.

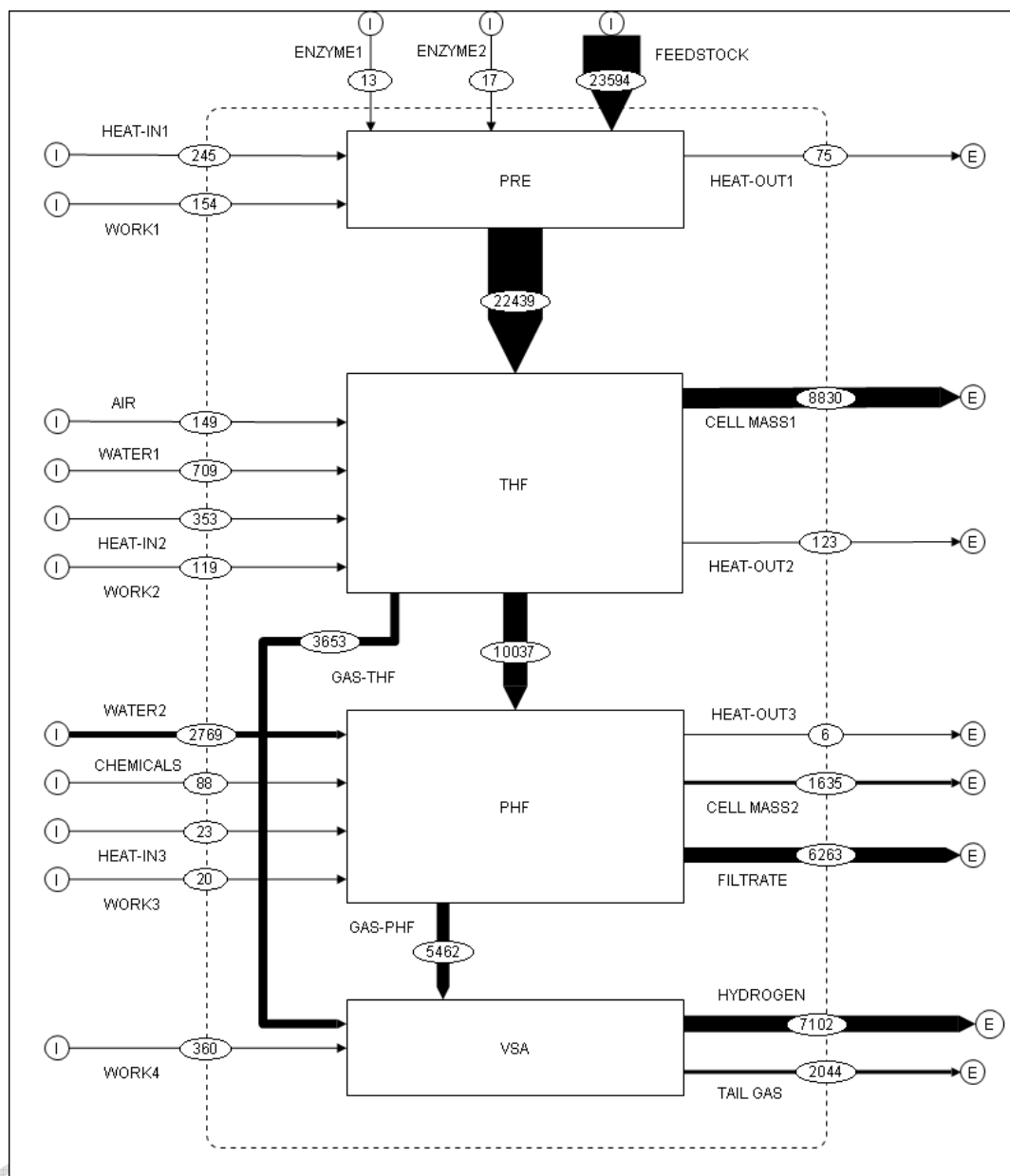


Fig. 2. Exergy balance of process option without stripping; gas-upgrading by VSA [MJ/h].

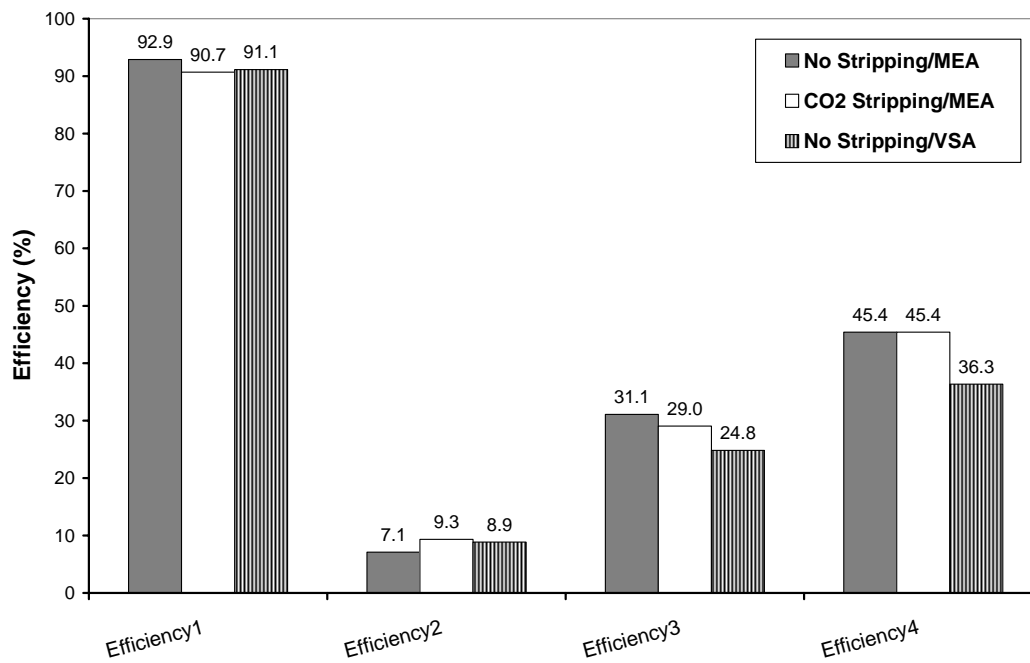


Fig. 3. Overall exergetic efficiency of investigated process options.

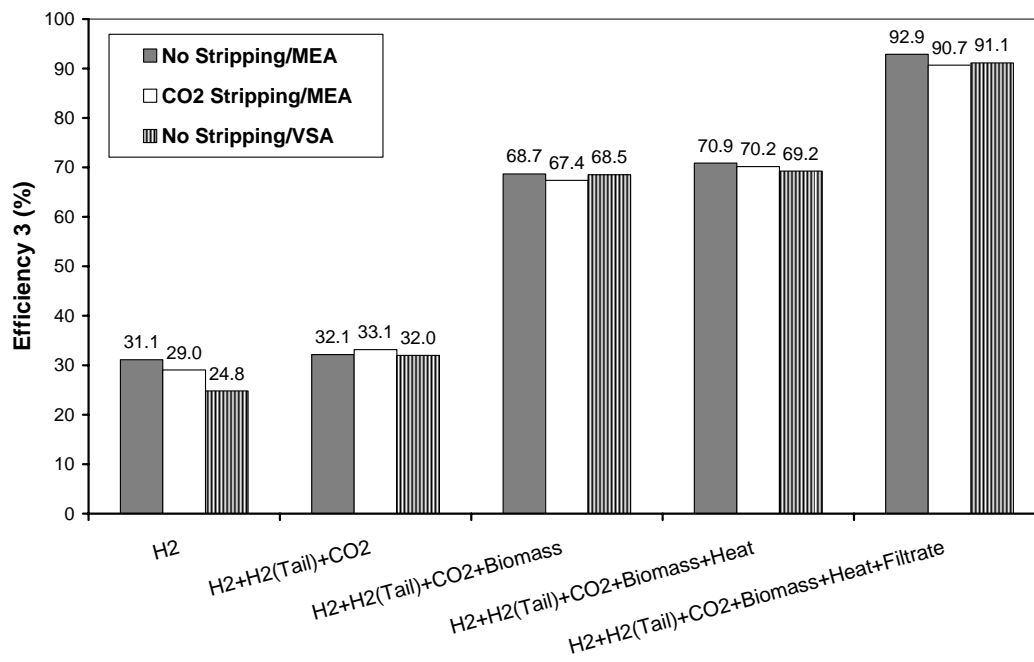


Fig. 4. Parameter study on product definition – Rational exergetic efficiency (Efficiency 3).

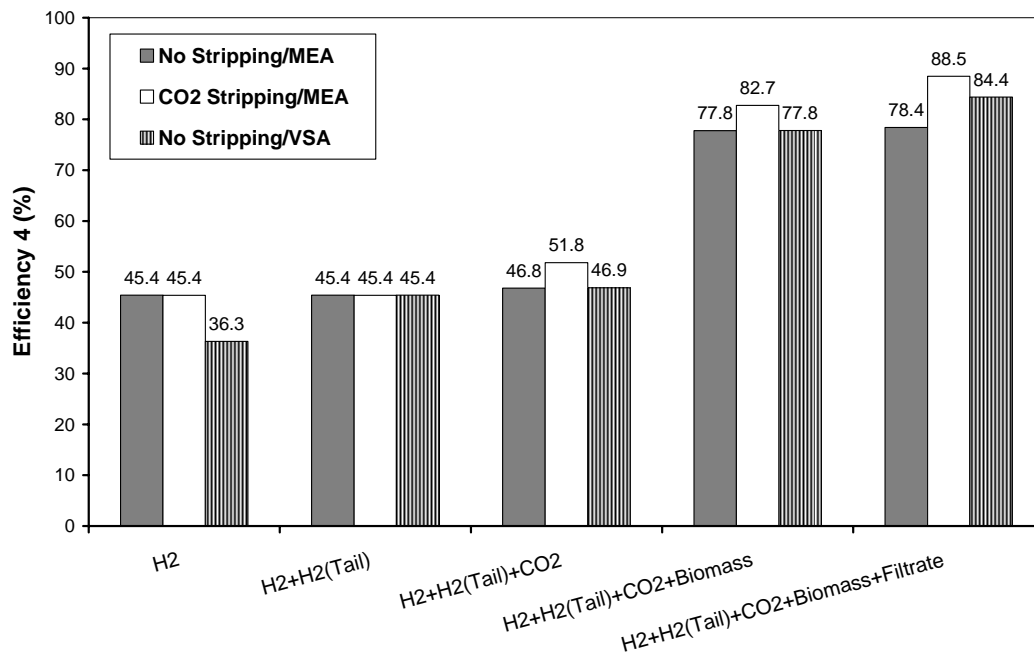


Fig. 5. Parameter study on product definition – Chemical exergetic efficiency (Efficiency 4).