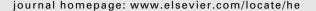
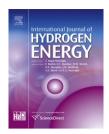


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H₂ processes with CO₂ mitigation: Thermo-economic modeling and process integration

Laurence Tock*, François Maréchal

Industrial Energy Systems Laboratory, Ecole Polytechnique Fédérale de Lausanne, Station 9, CH-1015 Lausanne, Switzerland

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ABSTRACT

Within the challenge of greenhouse gas reduction, hydrogen is regarded as a promising decarbonized energy vector. The hydrogen production by natural gas reforming and lignocellulosic biomass gasification are systematically analyzed by developing thermoeconomic models. Taking into account thermodynamic, economic and environmental factors, process options with CO_2 mitigation are compared and optimized by combining flowsheeting with process integration, economic analysis and life cycle assessment in a multi-objective optimization framework. The systems performance is improved by introducing process integration maximizing the heat recovery and valorizing the waste heat. Energy efficiencies up to 80% and production costs of $12.5-42~\text{s/GJ}_{\text{H}_2}$ are computed for natural gas H_2 processes compared to 60% and $29-61~\text{s/GJ}_{\text{H}_2}$ for biomass processes. Compared to processes without CO_2 mitigation, the CO_2 avoidance costs are in the range of $14-306~\text{s/t}_{CO_2,avoided}$. The study shows that the thermo-chemical H_2 production has to be analyzed as a polygeneration unit producing hydrogen, captured CO_2 , heat and electricity. Copyright © 2012, Hydrogen Energy Publications, LLC. Published by Elsevier Ltd. All rights reserved.

1. Introduction

Within the worldwide challenge of global warming mitigation and energy security, renewable resources and carbon capture and storage (CCS) have received considerable attention, especially for hydrogen and electricity production. Biomass-based processes [1] emitting no CO_2 if carefully managed and other renewable H_2 production processes [2] show a high potential toward a sustainable future. H_2 is regarded as a clean, reliable and affordable energy vector that can substitute fossil fuels by the combustion in an internal combustion engine or by electrochemical conversion to electricity in a fuel cell system with high efficiency and without on site CO_2 emissions. In this perspective, the pre-combustion or hydrogen routes are investigated here with regard to different resources (i.e. wood and natural gas) and competing outputs

such as H_2 and/or electricity and captured CO_2 , and their combination in polygeneration systems.

Several research studies have already identified promising fuel decarbonization processes for H₂ production and/ or electricity generation using different resources. As an example some H₂ process performance results are summarized in Table 1. Reported efficiencies range from 69 to 80% for fossil fuel H₂ production [3–5] and from 51 to 60% for biomass fed processes [6,7]. In each study, different assumptions are made and different technologies are considered. This yields a large range of performance results making a consistent comparison difficult. The reaction characteristics of H₂ production by reforming and partial oxidation of natural gas have been studied based on thermodynamic analysis in Refs. [8–10] and for biomass processes in Ref. [11]. In Ref. [12] the economics of producing

^{*} Corresponding author. Tel.: +41 21 693 3528; fax: +41 21 693 3502.

Nomenclature		TIT WGS	turbine inlet temperature water—gas shift
Abbrevio	autothermal reforming biomass carbon capture carbon capture and storage cold gas cleaning electricity import fast internally circulating fluidized bed gas turbine higher heating value high temperature shift life cycle assessment lower heating value low temperature shift methyldiethanolamine monoethanolamine mechanical vapor recompression natural gas natural gas combined cycle partial oxidation pressure swing adsorption self sufficient (in terms of energy) steam methane reforming triethanolamine	Greek le Δh ⁰ Δh̄ _r ε _{eq} ε _{tot} η _{CO₂} θ _{wood} Roman C È ṁ P Q T Subscrip cc ref res Superscrit -	lower heating value, kJ/kg standard heat of reaction at 25 °C, kJ/mol natural gas equivalent efficiency, % energy efficiency, % CO2 capture rate, % wood humidity, %wt letters production cost, \$/GJ mechanical/electrical power, kW mass flow, kg/s pressure, bar heat, kW temperature, K ots plant with carbon capture reference plant without carbon capture resource: natural gas (NG) or wood (BM)

 H_2 from fossil and renewable resources are compared; for natural gas fed processes producing 236–341 $t_{\rm H_2}/d$ costs of 19–27\$ $_{\rm 2007}/GJ_{\rm H_2}$ with a gas price of 10.3 \$/GJ_{NG} are reported. While for renewable processes using biomass, solar or wind to produce between 1.3 and 354 $t_{\rm H_2}/d$ costs in the range of 19.5–70.3 \$/GJ_{\rm H_2} are reported [12]. These costs values are however highly dependent on the resource prices that constitute about one to two third of the cost. These studies [4,12] comparing H_2 processes using various resources and technologies are mainly based on a literature survey with regard to the production cost and do not include process modeling and optimization.

The co-production of electricity and H_2 from natural gas resources is studied in Ref. [5] computing energy and exergy efficiencies to assess the benefit in terms of primary energy consumption and/or reduced CO_2 emissions. Besides performance estimates and thermodynamic arguments, no economic analysis is however performed. In Ref. [13] the thermodynamic and engineering aspects of pre-combustion natural gas power plants are studied without including

energy integration and economic aspects. However, in Ref. [14] it is shown how heat recovery for reactants preheating can increase the $\rm H_2$ yield by 10%. Whereas for coal based $\rm H_2$ and electricity co-production processes, the studies in Refs. [15,16] included efficiency and cost evaluations and [17] applied process integration to maximize the overall plant energy efficiency.

To overcome the difficulties of comparing processes with different assumptions, our goal is to propose a comprehensive comparison framework combining thermo-economic models, energy integration techniques and economic evaluation simultaneously. The objective is to compare and optimize fuel decarbonization (pre-combustion) process configurations with regard to energy, economic and environmental considerations by applying a consistent methodology. Special interest is given to the effect of polygeneration of H_2 fuel, captured CO_2 , heat and power, in order to identify its advantages and constraints, and to better understand trade-offs between efficiency, investment and emissions.

Process	CO ₂ capt. [%]	∈ [%]	$[\$/GJ_{H_2}]$	$[t_{H_2}/d]$	Res. price	Ref.
Natural gas	0	83.9 _(HHV)	5.2	418	3 \$/GJ _{NG}	[3]
Natural gas	71	78.6 _(HHV)	5.6	418	3 \$/GJ _{NG}	[3]
Coal (Texaco gasif.)	0	63.7 _(HHV)	8.7	309	29 \$/t	[12]
Coal (Texaco gasif.)	87	59 _(HHV)	10.5	281	29 \$/t	[12]
Biomass (FICFB, CGC)	-	57.7	_	-	-	[7]
Biomass	_	51-60	8-11	90-184	2 \$/GJ _{BM}	[6]

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