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Renewable hydrogen production concepts from bioethanol reforming with carbon capture



HYDROGEN

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ABSTRACT

This paper is assessing the hydrogen production from bioethanol at industrial scale (100000 Nm³/h hydrogen equivalent to 300 MW thermal) with carbon capture. Three carbon capture designs were investigated, one based on pre-combustion capture using chemical gas—liquid absorption and two based on chemical looping (one based on syngas and one using direct bioethanol looping). The carbon capture options were compared with the similar designs without carbon capture. The designs were simulated to produce mass and energy balances for quantification of key performance indicators. A particular accent is put on assessment of reforming technologies (steam and oxygen-blown autothermal reforming) and chemical looping units, process integration issues of carbon capture step within the plant, modelling and simulation of whole plant, thermal and power integration of various plant sub-systems by pinch analysis. The results for chemical looping designs (either syngas-based or direct bioethanol) show promising energy efficiency coupled with total carbon capture rate.

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1. Introduction

Deployment of hydrogen as new energy carrier complementary to electricity and conventional fossil fuels (e.g. natural gas, oil derived products, coal etc.) is raising much interest, as this offers significant advantages, e.g. no greenhouse gas emissions when combusted, security of energy supply, higher energy conversion efficiencies and better economic performances [1]. Currently, hydrogen is mostly used in petrochemical industry, e.g. ammonia and methanol synthesis, hydrogenation, hydrocracking and hydrodesulphurization. Hydrogen is produced mainly from fossil fuels (natural gas reforming and coal gasification) [2–5]. Developing high energy efficient hydrogen production methods based on renewable energy sources (e.g. solar, wind, biomass etc.) is an important factor to be considered in the hydrogen value chain and to develop hydrogen-based large scale applications [4,6,7]. Within this context of developing low carbon applications based on renewable energy sources and carbon capture and storage, it worth mention that European Commission has set targets for EU that until 2020, 20% from energy mix should be covered by renewable energy sources, 20% cut of CO_2 emissions compared with 1990 levels as well as 20% save of energy consumption by improving energy efficiency [8].

The developing of hydrogen applications for future lowcarbon economy imply for instance power generation using hydrogen-fuelled machines (e.g. gas turbine, fuel cells) and also transport applications using PEM fuel cells. The issues of environmental protection, combating climate change by

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reducing CO_2 emissions, enhancing the security of energy supply and improving economic competitiveness shape the future energy sector. In this context, developing new hydrogen production routes which use renewable sources with low or negative CO_2 emissions are of particular importance. Within this context, biomass conversion methods for energy vector poly-generation (including hydrogen) are targeted.

This paper evaluates various innovative hydrogen production options using bioethanol as feedstock simultaneous with carbon capture. There are several bioethanol reforming possibilities (catalytically conventional or authothermal reforming) for hydrogen production as well as carbon capture options (gas—liquid and gas—solid applications) to be integrated in the overall design. Bioethanol reforming for hydrogen production implies the following global reaction [7,9]:

$$C_2H_5OH + 3H_2O \rightarrow 2CO_2 + 6H_2 \quad \Delta H = +157 \text{ kJ/mole}$$
 (1)

Apart of the main chemical Reaction (1), other several side reactions could take places but the operational conditions are chosen in such a way to minimize their extent [10]. The major components in the syngas produced by reforming are hydrogen, carbon dioxide and carbon monoxide together with other minor components like methane, ethene, acetaldehyde etc. Two reforming technologies were investigated in the paper: the first design is conventional steam reforming in which the heat balance for Reaction (1) is covered by an external heater. The second design is an oxygen-blown reforming process in which oxygen is added to totally oxidize part of bioethanol to cover the reforming heat duty for the remaining fuel. In a design for hydrogen production, the carbon monoxide from the syngas is then converted with water according to the following chemical reaction (water gas shift - WGS):

$$CO + H_2O \rightarrow CO_2 + H_2$$
 $\Delta H = -41.02 \text{ kJ/mole}$ (2)

The shift conversion has a double purpose: to concentrate the syngas energy in form of hydrogen and to convert the carbon species into carbon dioxide. CO_2 can be later captured by gas—liquid physical or chemical absorption in a precombustion capture configuration [11–13]. After carbon capture, the hydrogen-rich gas is purified by Pressure Swing Adsorption (PSA) to the desired specification (>99.95% vol. hydrogen). PSA tail gas and additional hydrogen-rich gas are used in an external burner to cover the heat duty of the reforming Reaction (1).

Another promising carbon capture option to be integrated in energy conversion systems is based on chemical looping. The chemical looping method consists in two processes (oxidation and reduction) undertaken in separate reactors [14,15]. In the reduction step, the fuel (hydrocarbons, syngas or direct fuel) is reacted with an oxygen carrier (metallic oxide) to form carbon dioxide and water. This reactor (called fuel reactor) is operated mostly in fluidisation mode. After condensing the water vapour and conditioning (drying and compression), the captured carbon dioxide stream can be send to the storage sites. The reduced form (lower oxidation stage or even metal) of the oxygen carrier is then re-oxidised using steam and/or air and recycled back to the fuel reactor. For the particular case of iron oxide (haematite) evaluated in this paper as oxygen carrier, the reduce form (Fe/FeO) resulted from the fuel reactor is partially re-oxidised with steam to magnetite (Fe₃O₄) and hydrogen in the steam reactor. Then the solid phase is fully oxidised with air back to Fe₂O₃ in the air reactor. All gas streams from the chemical looping unit reactors are cooled down to generate steam. High temperature heat recovery potential is one distinct advantage of chemical looping systems to provide high energy efficiencies. Considering the syngas resulted from bioethanol catalytic reforming process, the chemical reactions are the following [16,17]:

- Fuel (syngas) reactor:

 $Fe_2O_3 + 3CO \rightarrow 2Fe + 3CO_2 \quad \Delta H = -12.0 \text{ kJ/mole}$ (3)

$$Fe_2O_3 + 3H_2 \rightarrow 2Fe + 3H_2O \quad \Delta H = +20.2 \text{ kJ/mole}$$
(4)

 $4Fe_2O_3 + 3CH_4 \rightarrow 8Fe + 3CO_2 + 6H_2O \quad \Delta H = +274.5 \text{ kJ/mole}$

- Steam reactor:

$$3Fe + 4H_2O \rightarrow Fe_3O_4 + 4H_2 \quad \Delta H = -150.8 \text{ kJ/mole}$$
 (6)

- Air reactor:

$$2Fe_3O_4 + 1/2O_2 \rightarrow 3Fe_2O_3 \quad \Delta H = -478.817 \text{ kJ/mole}$$
 (7)

Another chemical looping option evaluated in this paper is based on direct bioethanol conversion in the fuel reactor according to the Reaction (8). The reduced solid carrier resulted from fuel reactor is then processed in the steam and air reactors according to the Reactions (6) and (7) for hydrogen production and solid carrier re-oxidation.

$$2Fe_2O_3 + C_2H_5OH \to 4Fe + 2CO_2 + 3H_2O$$
 (8)

The following bioethanol-based plant configurations for hydrogen production with or without carbon capture were evaluated:

Case 1: Hydrogen production by bioethanol reforming with carbon capture using gas-liquid absorption (MDEA);

Case 2: Hydrogen production by bioethanol reforming with syngas-based iron cycle;

Case 3: Hydrogen production by direct bioethanol iron cycle;

Case 4: Hydrogen production by bioethanol reforming without carbon capture.

For Cases 1 and 4, both bioethanol reforming technologies were assessed (steam reforming – Case 1a/4a and oxygenblown authotermal reforming – Case 1b/4b). The bioethanol reforming without carbon capture (Cases 4a and 4b) were only considered as benchmark cases for carbon capture designs. For all evaluated cases, a hydrogen stream of 100000 Nm³/h (equivalent to 300 MW thermal considering hydrogen lower heating value) was produced. A specific accent was put in the paper on the evaluation of technical performances (e.g. fuel consumption, energy efficiency, ancillary consumption, Download English Version:

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