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Life Cycle Assessment of Hydrogen Production Processes: Steam Reforming of Natural Gas, Ethanol and Bioethanol

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1 Introduction

Nowadays, natural gas is the main raw material for obtaining hydrogen through steam reforming. One of the most important environmental constraints of this process is related to its high greenhouse gases emissions. The global warming potential of this process can reach 13.7 kg CO_2 (equiv.) per kilogram of net hydrogen produced [1]. A part of these emissions is inherent to the process since CO_2 is a co-product in the main reactions involved: reforming reaction ($CH_4 + H_2O \leftrightarrows CO + 3H_2$) and water gas shift reaction ($CO + H_2O \leftrightarrows CO_2 + H_2$). The rest of the emitted CO_2 is related to natural gas extraction, processing of materials necessary for the plant, ancillary materials and energy consumption (heating, compression…) during the process.

In order to decrease these greenhouse gases (GHG) emissions some alternatives such as the implementation of CO_2 capture and storage (CCS) techniques and the use of others raw materials for the reforming reaction are being evaluated. For instance, steam reforming of ethanol is an attractive route because its high hydrogen content, easy-handling and low toxicity [2]. However, if the origin of ethanol is from fossil fuel, problems of CO_2 emissions remain being also necessary the use of CCS techniques. On the contrary, the use of bioethanol, easily produced via fermentation of biomass or agricultural waste products, is probably the most attractive alternative since CO_2 produced during its burning may be consumed during biomass growth [2].

When developing novel alternatives for hydrogen production, not only the reduction of CO_2 emissions and technical aspects must be taken into account, but the total environmental impact has to be also considered [3]. The main issue should be to establish which is the most environmentally-friendly process from a global point of view. In this sense, Life Cycle Assessment (LCA) is a powerful tool since it takes into account all the stages in the processes for hydrogen production, from the construction materials needed to erect the plants to the management of wastes generated during the operation.

In this work, hydrogen production through steam reforming of gas natural, ethanol and bioethanol including CCS techniques has been studied from a life-cycle point of view to determine which shows the best environmental performance from an overall point of view.

2 Systems Definitions and LCA Assumptions

The first system evaluated consists on the coupling of the methane steam reforming with CO_2 capture and storage (system called M-SR). An usual steam reformer is considered, operating at 1,100-1,200 K and 30 bar with a supported nickel catalyst, assuming that a 85 % conversion is obtained. The water gas shift reaction is carried out in two fixed beds working at high (773 K) and low temperature (573 K), respectively, with intermediate cooling [4]. Regarding to CO_2 capture and storage, it is assumed that CO_2 emissions from reforming are scrubbed with monoethanolamine, stripped, pressurized (130 bar) and injected in depleted oil and gas reservoirs [5].

Regarding to ethanol and bioethanol steam reforming (Et-SR and BIO-SR), the same process was considered for both cases by using a supported nickel catalyst at 773 K. Differences between both LCA studies are related to the ethanol origin: typical ethylene hydration and fermentation of sugar beets molasses, respectively. Processes associated with the land use (soil cultivation, sowing, fertilization, pest and pathogen control, etc.) as well as machine infrastructure necessary for sugar beets production were included in the inventory of bioethanol reforming. Likewise, processing of sugar beets to molasses at refinery including the treatment of effluents was also taken into account. Assumptions for CO_2 capture and storage after ethanol or bioethanol reforming were the same previously described for methane steam reforming.

For the three LCA studies, the selected functional unit is 1 Nm^3 of hydrogen (99.99% purity). The life cycle assessment is focused on the raw material, energy acquisition and manufacturing stages, as distribution, use and end-of-life stages are supposed to be the same for the three processes. The construction materials needed to erect the production plants are also considered. In order to perform a correct inventory for the LCA, systems under study were simulated determining the inputs (raw materials and energy) and outputs (materials, wastes and emissions) for each plant. The assessment was carried out with SimaPro 7.1 software by using the eco-invent 2.0 database. The Eco-indicator 99 method was used for impacts classification and characterization. This method takes into account the environmental effects that damage the human health, ecosystems quality and natural resources such as greenhouse effect, ozone layer depletion, carcinogens and respiratory effects, radiation, eco-toxicity, acidification, eutrophication, land use and consumption of minerals and fossil resources.

3 Results and Discussion

Table 1 shows the results of LCA´s for the three systems by using the above mentioned Eco-indicator 99 method. According to this methodology, the greenhouse effect, ozone layer depletion, respiratory effects and radiation are expressed as DALY (Disability Life Years, effects on human health); the eco-toxicity as PAF (Potentially Affected Fraction, effect on ecosystem quality); the acidification, eutrophication and land use as PDF (Potentially Disappeared Fraction, effects on ecosystem quality) and the consumption of minerals and fossil resources as surplus energy necessary for future extractions of low quality minerals and fossil resources (MJ surplus, effects on available resources).

Regarding to the effects on human health of the three processes, it is very remarkable the reduction of climate change achieved by bioethanol reforming. This result indicates that biomass growth allows fixing more CO_2 than the produced in the subsequent processes. However, as can be seen in the Figure 1, others impacts that damage the human health such as respiratory effects and radiation are higher for steam reforming of bioethanol than for M-SR and Et-SR processes. The production and use of fertilizers and pesticides necessary for the cultivation step are probably responsible of these higher DALY values.

Table 1: LCA results by using Eco-indicator 99.

Impact category	**Unit**	**M-SR**	**Et-SR**	**BIO-SR**
Respiratory effects	DALY	2.637E-07	3.378E-07	4.140E-07
Climate change	DALY	1.023E-07	1.346E-07	-1.162E-07
Radiation	DALY	2.623E-09	2.890E-09	6.602E-09
Ozone layer	DALY	2.357E-10	1.555E-11	5.739E-11
Eco-toxicity	PAF*m^2yr	3.559E-02	1.169E-02	5.048E-02
Acidification/ Eutrophication	PDF*m^2yr	7.991E-03	9.708E-03	2.090E-02
Land use	PDF*m^2yr	6.762E-03	1.788E-03	5.327E-01
Minerals	MJ surplus	5.535E-03	4.075E-03	1.542E-02
Fossil fuels	MJ surplus	4.243	3.749	0.896

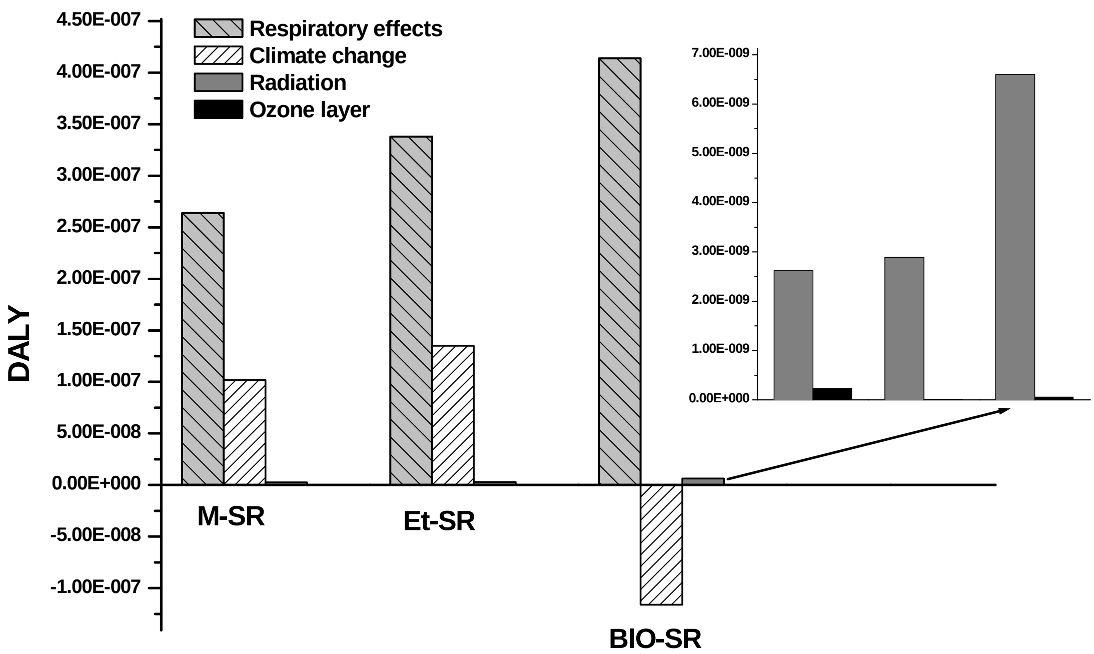

Figure 1: Effects on human health of M-SR, Et-SR and BIO-SR processes (Eco-Indicator 99).

Figure 2 shows the impacts on the ecosystem quality of the three processes indicating that bioethanol steam reforming leads to more acidification, eutrophication and eco-toxicity than M-SR and Et-SR. As known, the application of fertilizers and pesticides on the land for improving its cultivation capacity and controlling pathogen populations favours the acidification and eutrophication and increases the environment toxicity for a lot of species. Some authors propose that acidification is mainly caused by atmospheric NH_3 emissions coming from nitrogen of fertilizers [6] whereas others investigations suggest that emissions from diesel of agricultural machinery have also an important contribution to this effect [7]. Regarding to the eco-toxicity, results previously reported also indicate that it is widely increased by agricultural activities due to production of fertilizes and emissions of pesticides [7, 8].

Eco-indicator 99 methodology is recommended for the evaluation of processes which include cultivation stages since it takes into account an important penalty for the land use. In this case, it is obvious that environmental impact associated with the land use must be much higher for bioethanol reforming than for M-SR and Et-SR processes due to the biomass cultivation step.

The results obtained for the use of mineral and fossil resources are especially remarkable (see Table 1). Despite reforming of bioethanol consumes more minerals than the other two processes due to the utilization of fertilizers, it presents much lower fossil fuel consumption. These results are related to the raw materials used for each system: biomass for BIO-SR and methane and ethylene coming for the oil for M-SR and Et-SR, respectively, and the reuse of biomass wastes to produce energy.

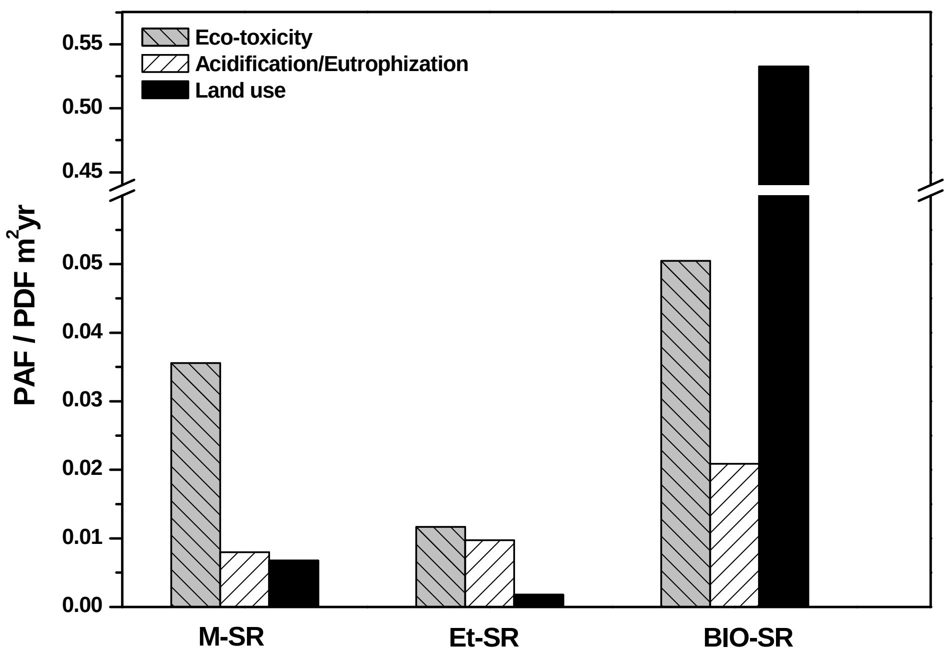

Figure 2: Effects on ecosystems quality of M-SR, Et-SR and BIO-SR processes (Eco-Indicator 99).

Finally, Figure 3 shows the single score for the three processes calculated according to Eco-indicator 99 methodology, after normalization (factors 65,4 for human health categories, $1.95 \cdot 10^{-4}$ for ecosystems quality categories and $1.19 \cdot 10^{-4}$ for resources ones) and weighting (400, 400 and 200, respectively). It can be observed that the most environmental-friendly process is the reforming of bioethanol. That means the high environmental impacts associated with the production and utilization of fertilizers and pesticides and land use are clearly compensated by the low consumption of fossil fuels and the capture of CO_2 during biomass growth.

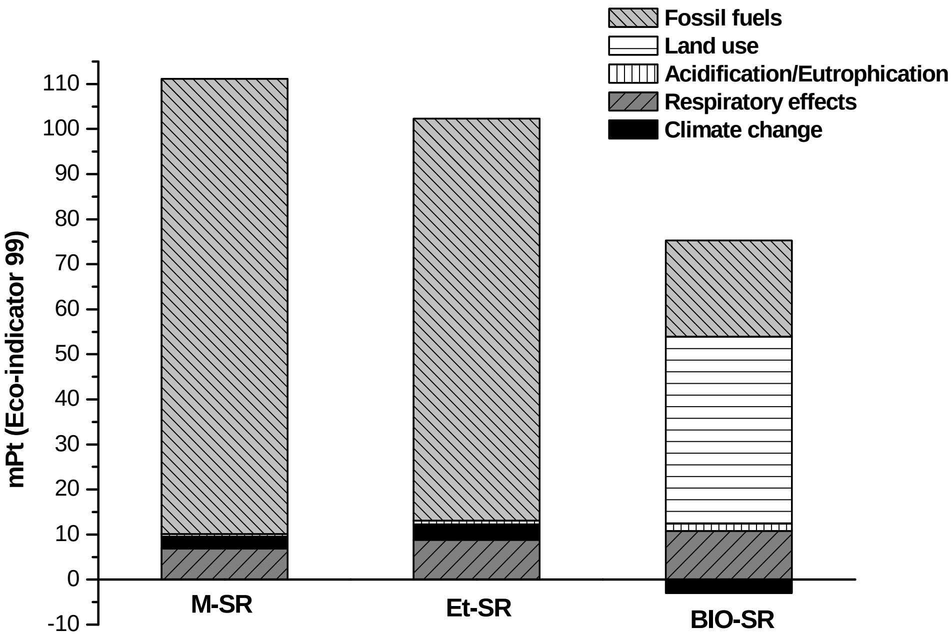

Figure 3: Single score of M-SR, Et-SR and BIO-SR processes (Eco-Indicator 99).

The conclusion that bioethanol steam reforming is the most environmental-friendly process from a global point of view is important since not always processes which lead to less greenhouse gases emissions also present the lowest single score. For instance, previous works have showed that the incorporation of CO_2 capture and storage techniques to the conventional methane steam reforming allows a great decrease of CO_2 emissions but leads to a slight increase of the process total single score (principally due to higher NOx emissions associated with the high electricity consumption) [3]. But in this case, even by using a methodology as Eco-indicator 99 which penalties the land use (and, therefore, the cultivation stages), the reforming of bioethanol seems to be the most environmental-friendly option for hydrogen production.

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