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Life Cycle Assessment of Simulated Hydrogen Production by Methane Steam Reforming

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ABSTRACT

Hydrogen has attracted global attention as alternative energy carrier in the future. Typically, hydrogen is produced through methane steam reforming (MSR) followed by water gas shift (WGS) reaction. Although considered as clean energy, it is essential to assess the environmental impact of hydrogen production process which could help to compare and improve existing technology. Thus, the objective of this study is to conduct a life cycle assessment (LCA) of hydrogen production from natural gas (NG) as feedstock. In order to gain detail and extensive process inventory, a rigorous flowsheet simulation of hydrogen production was developed in Aspen Plus 8.6. The goal of LCA is to evaluate the environmental impact of all processes involved in hydrogen production from natural gas. The environmental assessment was carried out using GaBi based on ReCiPe method. The system boundaries considered for this assessment were natural gas feedstock, hydrogen production, process steam, process water plant and solvent absorption. The LCA system function is the production of hydrogen from methane while the functional unit chosen is 1 kg of hydrogen. Overall, ten life cycle impact assessment categories were carried out. Our findings show that the most contributing impact categories were climate change and resource depletion which include fossil and water.

INTRODUCTION

Energy resources is important to satisfy human needs. However, excessive exploitation of energy resources could lead to crucial environmental consequences. Worldwide uncertainty in energy supply, the increasing oil price and the level of greenhouse gas emission have motivated the researcher to find new energy source to reduce dependence on non-renewable sources such as fossil fuels (Lee *et al*., 2010). In many countries, proactive actions have been taken to reduce the greenhouse gas (GHG) emissions in the energy sector (Tonini & Astrup, 2012). Hydrogen has been proposed as one of the future energy carriers because its high yields, clean combustion and feasible storage (Javier Dufour *et al*., 2011). Hydrogen mostly produced from natural gas via methane steam reforming (MSR) followed by water gas shift (WGS). It is the most widely method used in industries for the last 20 years (Tugnoli *et al*., 2008). While energy demand increases with increasing world population, hydrogen although considered as clean combustion gas could cause significant environmental impact due to increase greenhouse gas released during its production stage. In order, to assess the environmental impact, life cycle assessment (LCA) is a suitable tool to assess and compare the environmental impact of

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hydrogen production as it able fully evaluate the environmental impact from initial raw material until the final product.

LCA represents a systematic set of procedures for compiling and inspecting the inputs and outputs of materials and energy and the related environmental impacts and directly attributable to a product or service throughout its life cycle (Kalinci *et al*., 2012). LCA have been widely used in various industries such as paper production, car manufacturing and many more. For hydrogen production, several researchers adopted LCA at different stage of its product life cycle such as production, storage, transport and usage (J. Dufour *et al*., 2012). Dufour *et al*. (2012) for example perform LCA of various hydrogen production technology to determine which has the lower amount of greenhouse gas and total impact on the environment. In another work, Hajjaji *et al*.,(2013) performed LCA on various alternatives of hydrogen production from numerous feedstock. In the same study, the reactor models used to simulate the hydrogen production in the Aspen Plus was the equilibrium reactor. In this work, a rigorous model was used to simulate the hydrogen production in Aspen Plus 8.6 which include reaction kinetics, separation equilibrium models, production capacity and utility consumptions. Based on the simulated model, LCA analysis were the performed which will ensure a detail and extensive life cycle inventory for an accurate LCA results. The latest LCA method have been adopted in this work known as Recipe, which is an improvement from CML2000 and Eco-Indicator-99 (Consultants, 2016). The boundary of analysis considered in this work was gate to cradle which involve natural gas feedstock, hydrogen production, process steam, process water plant and solvent absorption. LCA analysis was done by using GaBi software for data collection, analyse and monitor the environmental performance of the process.

Methodology:
Hydrogen Production from Methane:

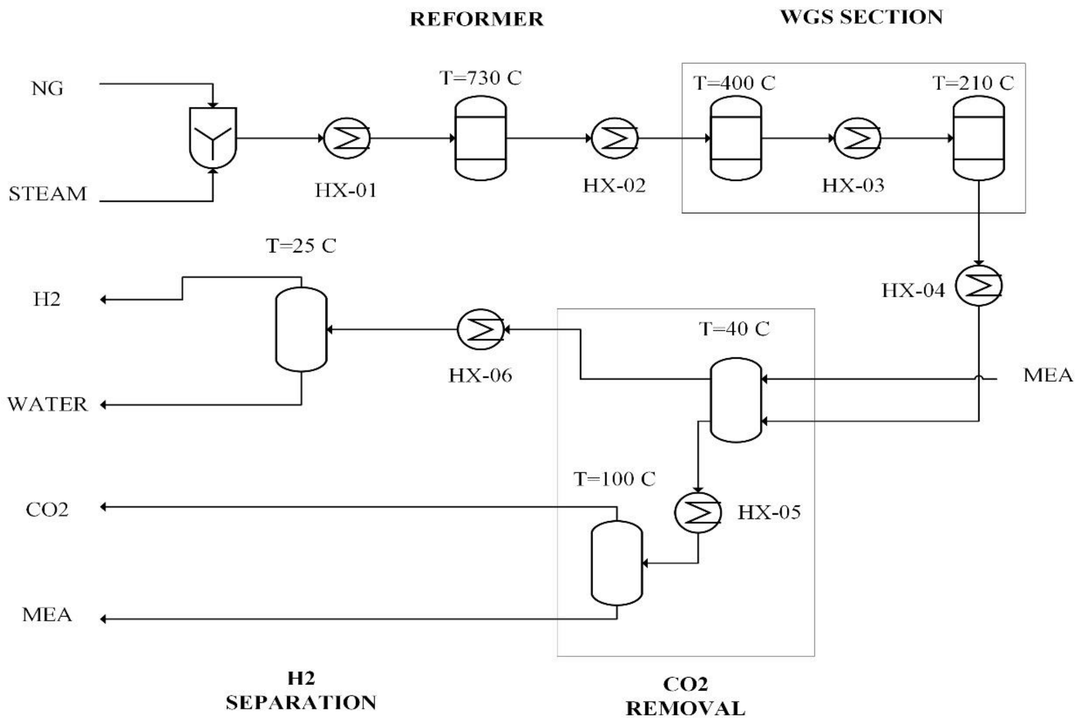

Fig. 1: The simplified flow sheet of hydrogen production by MSR.

Figure 1 shows a simplified flow sheet of hydrogen production by MSR reaction in the reformer followed byWGS reaction. The feed streams contain steam and methane from natural gas. Before entering the reformer, both streams were preheated to 730 C using heat exchanger. The MSR reaction products from the reformer was a mixture of CO, H_2, CH_4 and H_2O according to the following reaction:-

$$CH_4 + H_2O \rightarrow CO + 3H_2 \quad (1)$$

Then, the reaction products enters two adiabatic WGS reactors connected in series to increase the hydrogen yield. The first WGS reactor operated at high temperature around 400 C while the latter operated at low temperature around 210 C. The reaction occurred based on the following equation:-

$$CO + H_2O \rightarrow CO_2 + H_2 \quad (2)$$

In order to obtain high purity of hydrogen, CO2 need to be removed from the system. Absorption column is commonly used using MEA as the absorpbent with efficiency up to 99%. Then, hydrogen were separated from water using a separator at temperature 25 C to achieve up to 93% purity. The pure hydrogen were then stored in a pressurized tank.

LCA Goal and Scope:

In LCA goal and scope step it is important to define the objective of the analysis, functional unit (FU) and system boundary. The goal of this study is to evaluate the environmental impact of all processes involved in hydrogen production from natural gas. The functional unit (FU) provide a basis for calculating the inputs and outputs. In this work, a common FU of 1 kg of hydrogen produced was selected (Galera & Gutiérrez Ortiz, 2015; Verma & Kumar, 2015). The system boundaries on the other hand, determined the process units to be included within the evaluated system. The system boundaries for this system is shown in Figure 2. It is a cradle-to-grave approach which starts from methane feedstock until hydrogen storage. In detail, the system boundaries consist of five subsystems namely methane feedstock (SB1), hydrogen production (SB2), process steam (SB3), solvent absorption (SB4) and process water plant (SB5). Note that, the construction and commissioning phases as well as energy consumptions were excluded from the analysis and will be our future work.

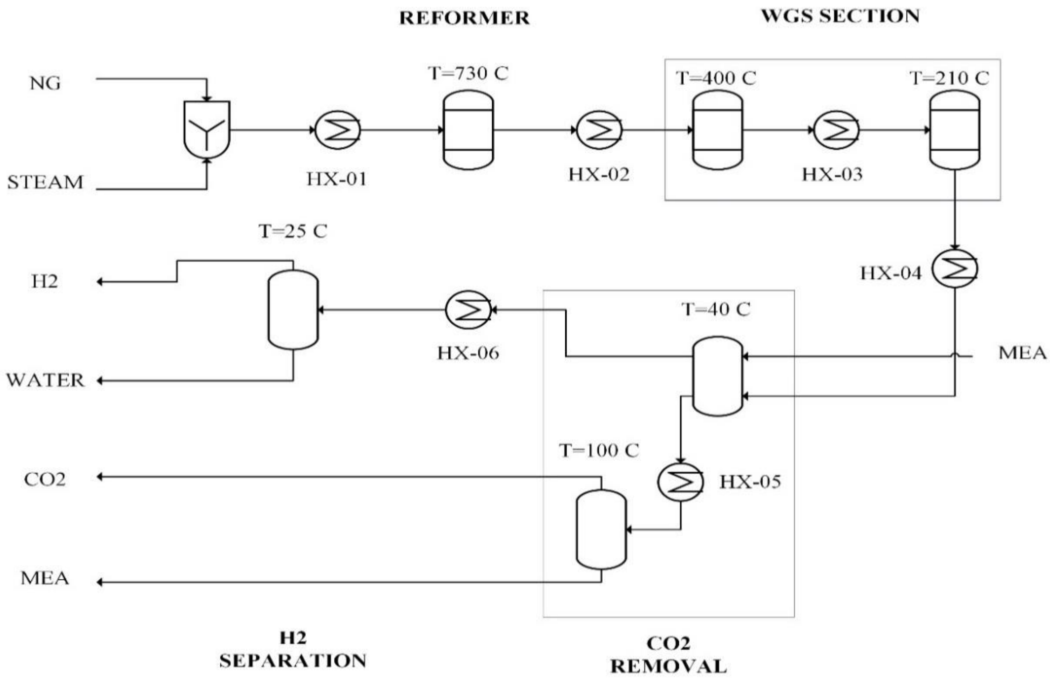

Fig. 1: The simplified flow sheet of hydrogen production by MSR.

For SB, the methane is a product of natural gas processing plant, its associated environmental impact was included in the analysis. However, the transportation of natural gas was assumed using pipeline and thus excludes from the analysis. SB2 consist of reactions and purification section. The reactions system includes a MSR and WGS reactor. In this section, methane reacts with steam to produce hydrogen in a MSR reactor while the gas produced then enters a WGS reactor to convert CO to CO_2 and increase hydrogen yield. The separation section on the other hand, consists of carbon dioxide removal and a separator. The aim of this section is to purify the hydrogen especially from carbon dioxide. The system boundary also considers process steam generation section (SB3). This section considers the combustion of hydrocarbon fuel in the boiler to generate steam which it used during plant operation. Meanwhile, SB4 is the MEA supply subsystem which supply absorbents for CO2 removal in the separation process. Finally, the water supply for the reforming process and cooling water were came from the process water plant (SB5).

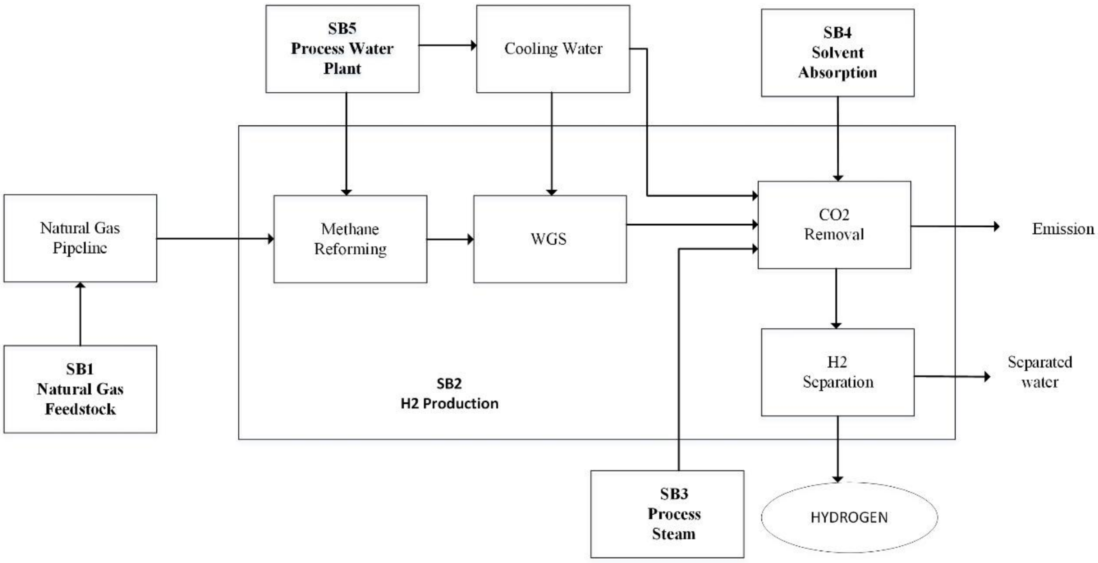

Fig. 2: System boundaries

Life cycle inventory (LCI):

LCI involves the collection and compilation of the data required to quantify all of the relevant inputs and outputs associated with the production of the functional unit (FU). In this study, Aspen Plus software were used to solve the mass and energy balances in hydrogen production from natural gas. The compounds used in this simulation includes hydrogen, carbon dioxide, carbon monoxide, methane, water and monoethanolamine (MEA). Figure 3 shows the flowsheet developed in Aspen Plus 8.6. The global thermodynamic method used in this simulation is electrolyte non-random two-liquid model Redlich Kwong (ENTRL-RK). Whereas, for MSR and WGS reactions the Redlich-Kwong-Soave Modified-Huron-Vidal mixing rule (RKSMHV2) were selected. This method is suitable for the mixture of non-polar and polar compound in combination with light gases. The process flowsheet is shown in Figure 3. For modelling the MSR and WGS reactions, RPLUG reactor block based on LHHW kinetics were selected. Whereas for the separation unit RADFRAC block model were selected for both absorption and stripper unit. Table 1 shows the specification of the models used in the simulation. The stream result summary is shown in the Table 2.

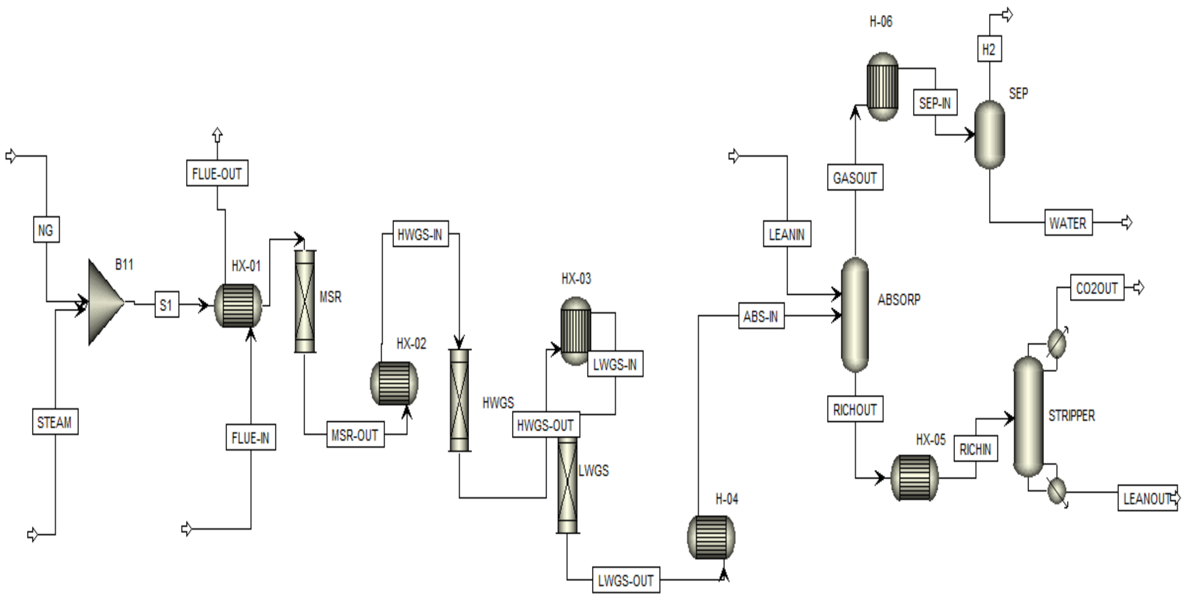

Fig. 3: Hydrogen production from methane flow-sheet in Aspen Plus.

Table 1: Summary of models and utilities used in the simulation.

Code	Equipment	Specification
HX-01	Heat exchanger (Heating)	Cold stream outlet temperature: 730 C Utility: Flue Gas
HX-02	Heat exchanger (Cooling)	Hot stream outlet temperature: 400 C Utility: Cooling water
HX-03	Heat exchanger (Cooling)	Hot stream outlet temperature: 210 C Utility: Cooling water
HX-04	Heat exchanger (Cooling)	Hot stream outlet temperature: 40 C Utility: Cooling water
HX-05	Heat exchanger (Heating)	Cold stream outlet temperature: 105 C Utility: Steam
HX-06	Heat exchanger (Cooling)	Hot stream outlet temperature: 28 C Utility: cooling water
MSR	reforming reactor	Operating temperature: 730 C Isothermal reactor
HWGS	water gas shift reactor	Operating temperature: 400 C RPLUG model block Adiabatic reactor
LWGS	water gas shift reactor	Operating temperature: 210 C RPLUG model block Adiabatic reactor
ABSORP	Absorption column	Number of stages:20 RADFRAC model block Packing size: 4X Packing material: Metal
STRIPPER	Stripper column	Number of stages: 20 RADFRAC model block Reboiler duty :5500 kW
SEP	Separator	Operating temperature: 25 C
Cooling water	Utility	Tin: 20 C / Tout: 40 C Pin: 1 atm / Pout: 1 atm
HP Steam	Utility	Tin: 250 C / Tout: 200 C Pin: 39 bar / Pin: 29 bar
Flue gas	Utility	Tin:1000 C / Tout: 792C Pin: 2 bar / Pout: 2 bar

Life cycle impact assessment (LCIA):

The life cycle impact assessment aims at understanding and evaluating the magnitude and significance of the potential environmental impacts of a product system throughout the life cycle of the product (ISO, 2006). The environmental characterization of the process was carried out based on the following categories; climate change, terrestrial acidification, fossil depletion, freshwater ecotoxicity, marine ecotoxicity, metal depletion, particulate matter, photochemical oxidant formation, water depletion and terrestrial ecotoxicity. The impact potentials were evaluated using ReCipe whereas the calculation implementation of the inventories was performed in GaBi.

Life Cycle Results Interpretation:

The data obtained from Aspen Plus were used as the main inventory data in GaBi. Table 3 summarize the main inventory data per functional unit of 1 kg hydrogen. Data for background processes such as natural gas or methane, steam and cooling water were taken from the GaBi database. The considered set of environmental impacts potential according to the latest LCIA method, ReCipe were climate change, terrestrial acidification, fossil depletion, freshwater ecotoxicity, marine ecotoxicity, metal depletion, particulate matter, photochemical oxidant formation, water depletion and terrestrial ecotoxicity. The results offers insights of the subsystems contributions to the environment impacts.

Table 2: Summary of stream results from Aspen Plus.

Mass Flow (kg/hr)	NG	STEAM	H2	CO2OUT	LEANIN	HWGS-OUT	WATER
MEA	0	0	9.13E-05	0.9523	18437.47	0	0.157741
H2O	0	8000	385.21	13281.12	1.10E+05	3423.63	2189.56
CO2	0	0	503.12	5348.10	0.0441563	4191.89	0.05541
OH-	0	0	0	0	0.4086	0	4.75E-05
HCO3-	0	0	0	0	222.19	0	4.1626
CO3-2	0	0	0	0	265.96	0	0.0345
MEAH+	0	0	0	0	12156.12	0	4.4052
MEACOO-	0	0	0	0	19073.73	0	0.1639
CO	0	0	329.32	0.1330	0	1779.48	1.07E-03
H2	3.23	0	1259.31	0.5196	0	1155.48	3.34E-03
CH4	2546.77	0	0	0	0	0	0

Total Flow	2550.00	8000.00	2476.96	18630.82	160404.00	10550.47	2198.55

Table 3: Main inventory data for the MSR system per functional unit

INPUT	Value	Unit
Flue gas	2.4	kg
Hydrogen	0	kg
Iron	0.504	kg
Monoethanolamine	14.75	kg
Natural gas	2.04	kg
Nickel	0.128	kg
Steam (MJ)	24.50	MJ
Steam superheated (hp)	6.4	kg
Water	88.20	kg
Water (cooling water)	386	kg
OUTPUT	Value	Unit
Carbon dioxide	5.18	kg
Carbon monoxide	0.2632	kg
Catalysts material	0.632	kg
Flue gas	2.4	kg
Hydrogen	1.00	kg
Monoethanolamine	14.75	kg
Waste water	90.07	kg

RESULTS AND DISCUSSIONS

The result for all environmental impacts potential for each subsystem is shown in the Table 4. Table 4 also shows the specific contribution of each subsystem for all impact categories impacts. Based on Table 4, the climate change category impact is the most significant impact to the environment followed by fossil depletion and water depletion. The other categories have minor environmental impact. The climate change also known as global warming potential (GWP) and resources depletion be worthy of discussions. The climate change quantifies the contribution of gaseous emission from the system to the environmental which include combination of CO_2, CH_4 and N_2O emissions. Figure 4 shows the variation of CO_2 emission in different system boundary. Hydrogen production subsystem contributed the most to climate change with 5.18 kg CO_2-eq per FU. This is because CO_2 was produced in the rectors as a side product. The CO_2 was removed by absorption column and emitted to the atmospheric. This is an agreement with the work by (Galera & Gutiérrez Ortiz, 2015; Hajjaji *et al*., 2013). Storing the CO_2 into liquid form is an option to reduce the greenhouse emission however it requires extensive energy for CO_2 liquification. Next after hydrogen production system is the process steam subsystem with 3.3 kg CO_2-eq per FU. This comes from burning of fossil fuels to generate high temperature steam. The effect of methane feedstock on climate change impact is low because the methane feedstock comes from natural gas, it contains high purity of methane and no carbon dioxide.

Table 4: Impact categories for each subsystems the hydrogen production from methane

Category	Unit	NG feedstock	Hydrogen production	Process steam	solvent absorption	Process water plant
Climate change	kg CO2-eq	1.29	5.18	3.3	0	1.48
Terestrial Acidification	kg SO2 -eq	1.77E-03	0	3.66E-03	0	3.16E-03
Fossil depletion	kg oil eq	2.694	0	1.403	0	0.505
freshwater ecotoxity	kg 1,4-DB eq	0	0	0	0.075	0.005
marine ecotoxiicity	kg N eq	9.30E-04	0	2.07E-03	0	4.66E-04
metal depletion	kg Fe eq	0.04	0.504	0.002	0	0.007
Particulate matter	kg PM10	0.000671	0	0.001416	0	1.07E03
Photochemical oxidant formation	kg NMVOC	0.003	0.012	0.006	0	0.003
Water depletation	m3	0.03	0.09	0.28	0	3.81
Terestrial ecotoxicity	kg 1,4-DB eq	0	0	0	0.212	0

Water depletion is defined as the net reduction in the availability of freshwater in a watershed for a given time period. Water depletion also reduces the water availability for current users and generating competition for the water resources. The water depletion can reduce resources availability for future generation and the intensity of the competition for freshwater will potentially increase. Based on water depletion impact in Figure 4, the process water plant subsystem contributes the most to water depletion impact categories compared to the other subsystem. This is obvious since water were used as a raw material to generate steam and also used as cooling water.

For consumption of fossil resources from Figure 4, it shows the natural gas feedstock contributes the most for fossil depletion impact category followed by steam production. This is expected since natural gas is a type of fossil fuel. The natural gas was used as feedstock in hydrogen production to react with steam and produce the hydrogen. In process steam, the natural gas was used as raw material to generate the steam to transfer the heat energy for heating in the system. So, the used fossil fuel as a raw material is giving the impact to fossil resources and it possible to happened the reduction of availability of fossil resource in the future. The future generation will be competing to get the fossil resource caused by the fossil depletion.

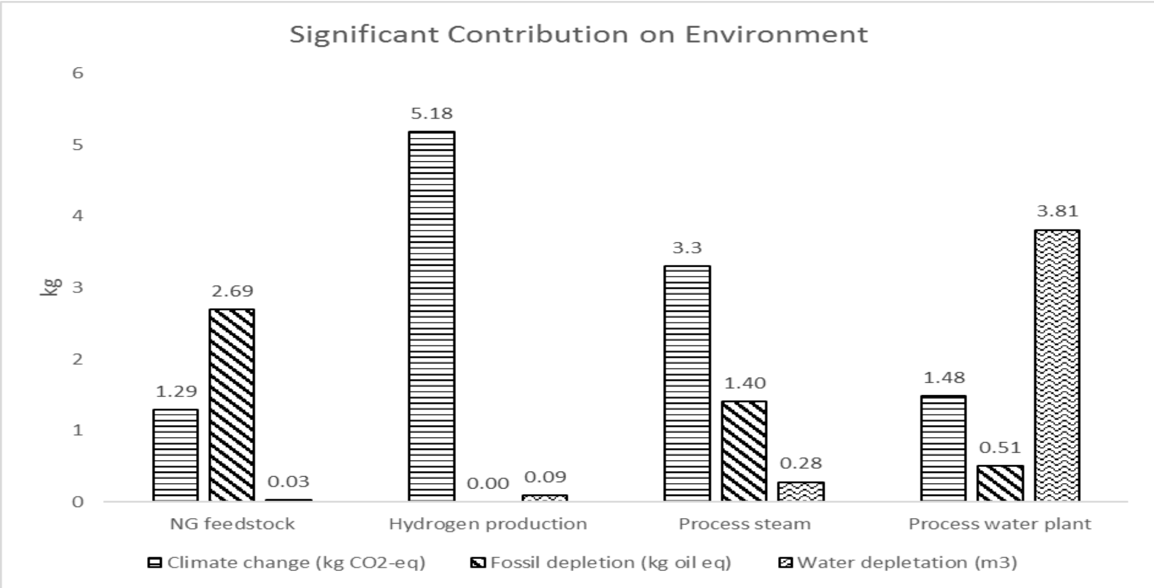

Fig. 4: Environmental impact results for the most significant impact category

Conclusion:

This work presents a life cycle assessment of a simulated hydrogen production from natural gas. Simulated model provide a detail and extensive data for conducting life cycle impact assessment. From the results obtained, the hydrogen production subsystem contributes the most to climate change impact category. Meanwhile, process water plant subsystem and natural gas feedstock subsystem contributes the most to water depletion and natural fossil depletion impact category respectively. The other impact categories just the minor significant in the environment impact. The life cycle approach is an excellent tool which can help identifying subsystems that contribute to the potential impacts attributes. Moreover, LCA could help to make decisions and improve process in reducing its impact to the environment.

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