

Environmental impact assessment of nuclear assisted hydrogen production via Cu–Cl thermochemical cycles

Ahmet Ozbilen*, Ibrahim Dincer, Marc A. Rosen

Faculty of Engineering and Applied Science, University of Ontario Institute of Technology, 2000 Simcoe Street North, Oshawa, Ontario L1H 7K4, Canada

ARTICLE INFO

Keywords:

Environmental impact
Thermochemical hydrogen production
Nuclear energy

ABSTRACT

The variation of environmental impacts with the efficiency of a hydrogen plant and the thermal output of a nuclear plant are studied for nuclear-based hydrogen production using three-, four- and five-step Cu–Cl cycles for thermochemical water splitting (TWS). Results are presented, on the basis of 1 kg hydrogen production, for two impact categories: global warming potential (GWP) and acidification potential (AP). Environmental impacts are evaluated with several approaches. First, environmental effects are calculated by varying the thermal output ratio, which represents the thermal energy used in Cu–Cl cycle divided by the total thermal energy output of nuclear plant, from 0.1 to 1. The results show that GWP can be decreased from 3.32 to 0.346 kg CO₂-eq for the five-step cycle. Second, the hydrogen plant efficiency is altered from 0.34 to 0.65 to examine the corresponding change in environmental impacts. Increasing the hydrogen plant efficiency to 0.65 decreases the GWP to 0.4 kg CO₂-eq and the AP to 2.1×10^{-3} kg SO₂-eq.

© 2012 Elsevier B.V. All rights reserved.

1. Introduction

The world's energy system, which is mainly based on fossil fuels, cannot be considered as sustainable. Concerns related to economic and environmental aspects of energy have been growing, particularly subsequent to the Kyoto Protocol of 1997 and the Stern Review of 2006. The risk of global climate change is of great concern to policymakers and to the public. The relation between the energy generation sector and environmental pollution is being carefully considered in industrialized and non-industrialized countries (Dincer & Balta, 2011). The International Energy Agency reports that the portion of global energy supplied by fossil fuels is 81.3% (IEA, 2010). Petroleum is particularly problematic, accounting for more than 95% of the energy consumption in the transportation sector. Economic and political implications of possible fossil-fuel shortages and environmental concerns have led to the search of alternative and cleaner energy technologies (Urbaniec, Friedl, Huisingsh, & Claassen, 2010). Although addressing future energy challenges requires numerous measures and approaches, the energy carrier hydrogen is expected by many to play a major role due to its emitting no greenhouse gases during oxidation. In addition, hydrogen may become widely used as a zero-CO₂ energy carrier for

vehicles and distributed heat and power generation using fuel cells (Davison, Arienti, Cotone, & Mancuso, 2010).

Hydrogen can be produced via many processes including as steam methane reforming, coal or biomass gasification, water electrolysis, and thermochemical water splitting (TWS), using such energy sources as fossil fuels, renewable energy and nuclear energy. The environmental impact associated with hydrogen use is based in large part on the production method. TWS splits water into hydrogen and oxygen through a series of thermally driven chemical reactions, and has the potential to be an environmentally benign and a cost-effective hydrogen production method. Only a few of the many reported TWS cycles (Funk, 2001) have progressed to experimental demonstrations. The copper–chlorine cycle is a promising TWS option, especially because of its lower temperature requirements (no higher than 530 °C).

Atomic Energy of Canada Limited (AECL) proposes linking nuclear energy with a hydrogen plant using a Cu–Cl TWS cycle because nuclear reactors (i) do not emit greenhouse gases during operation and (ii) are suitable for large-scale production. The Generation IV SCWR (super-critical water cooled reactor) is a particularly suitable option for pairing with the Cu–Cl TWS cycle.

Although hydrogen is a clean energy carrier, negative environmental impacts are associated with its production and merit investigation. Life cycle assessment (LCA) is useful tool for this task, since it provides an understanding of the environmental impacts of a good and/or a process, and potential reduction measures. In this study, LCA is used to evaluate the potential benefits of thermal management within the hydrogen production system and ways to

* Corresponding author. Tel.: +1 2899288918.

E-mail addresses: Ahmet.Ozbilen@uoit.ca (A. Ozbilen), Ibrahim.Dincer@uoit.ca (I. Dincer), Marc.Rosen@uoit.ca (M.A. Rosen).

Nomenclature

AP	acidification potential
B_d	discharge burn-up (GJ/kg U)
GWP	global warming potential
hrf	heat recovery fraction
LHV	lower heating value
η_{cyc}	hydrogen plant efficiency
$Q_{endo,cyc}$	reaction heat for endothermic reactions in the Cu–Cl cycle (MJ)
$Q_{exo,cyc}$	heat released by exothermic reactions (MJ)
$Q_{th,cyc}$	thermal energy requirement of the Cu–Cl cycle (MJ)
$Q_{th,nuc}$	thermal energy from nuclear plant (MJ)
Q_{waste}	waste heat from nuclear plant (MJ)
tor	thermal output ratio
$W_{el,cyc}$	electrical energy requirement of Cu–Cl cycle (MJ)
$W_{el,grid}$	electrical energy transferred to grid (MJ)
$W_{el,nuc}$	electrical energy from nuclear plant (MJ)

Acronyms

LCA	life cycle assessment
LCI	life cycle inventory
LCIA	life cycle impact assessment
NBHP	nuclear-based hydrogen production
SCWR	super-critical water cooled reactor
TWS	thermochemical water splitting

reduce environmental impacts. The specific objectives are to determine (1) the effect of thermal management measures that decrease the waste heat from the nuclear plant, and (2) the effect of hydrogen plant efficiency on two environmental impacts (GWP and AP).

2. Life cycle assessment

LCA is a useful tool for analyzing the environmental impacts associated with goods and services and for identifying opportunities for reducing the impacts attributable to wastes and resource consumption. All stages of a product or process (production, transportation, usage and disposal) are considered in the assessment. The four main phases of LCA (goal and scope definition, life cycle inventory, life cycle impact assessment and improvement analysis) are shown in Fig. 1, where arrows indicate that the phases are linked, and where an interpretation step is linked to all phases.

- Goal and scope definition** (ISO 14041, 1998): The first phase of the LCA clarifies respectively the results sought from the LCA, and the system considered and its boundaries. A functional unit is specified to provide a reference for the inputs and outputs.
- Life cycle inventory** (LCI) (ISO 14040, 1997): Life cycle inventory determines the environmental interventions (resource extractions from or emissions to an environmental compartment)

caused by or required for the processes within the boundaries of the studied system (Koornneef, van Keulen, Faaij, & Turkenburg, 2008). LCI requires identification of energy and material inputs and outputs for all flows across and within the system boundary for all processes, using data collection and calculation procedures. Obtaining data for LCI is often difficult (Curran, 2000).

- Life cycle impact assessment** (LCIA) (ISO 14042, 2000): Evaluation of environmental impacts of the material and energy flows, which are identified in the second phase of the LCA, is the aim of the LCIA. LCIA is divided into three steps (ISO 14042, 2000): (a) classification of impact categories and the assignment of inventory data to the categories; (b) characterization of inventory data within impact categories by quantifying their contributions to the impact categories; and (c) normalization and weighting to merge environmental impacts, if possible, and reduce them to fewer measures. A number of impact categories, characterization methods and factors for numerous substances are described by the Center of Environmental Science of Leiden University (CML), which is very useful for LCIA (Guinée et al., 2002).
- Improvement analysis**: Improvement analysis extends the LCI and LCIA results to develop potential improvements, in line with the goal and scope. Life cycle interpretation can assist in identifying and choosing the preferred alternatives, accounting for many factors (e.g. technical, economic and social).

In this paper, a unit product (1 kg of hydrogen) is considered. Also, two impact categories are considered in the LCIA phase:

- Global warming potential (GWP), which is defined as the impact of human emissions on the radiative forcing (i.e. thermal radiation absorption) of the atmosphere, and leads to climate change, which may affect ecosystem and human health. The earth's surface temperature is increased by emissions of greenhouse gases, which enhance radiative forcing (the "greenhouse effect"). GWP is measured in units of kg CO₂-eq.
- Acidification potential (AP), which is related to the deposition of acidifying pollutants on surface waters, ground water, soil, ecosystems, materials and biological organisms, and affects the natural environment, the anthropogenic environment, human health and natural resources. Major contributors to AP, which is measured in units of kg SO₂-eq, are SO₂, NO_x and NH_x.

3. Review of LCA of hydrogen production processes

LCAs have been reported for several TWS methods for hydrogen production. The net reaction for all processes for TWS is



Utgikar and Ward (2006) presented a LCA of a nuclear-assisted ISPR Mark 9 TWS cycle, a three-step thermochemical cycle involving iron chlorides (Fe–Cl). GWP and AP of the nuclear-based hydrogen production (NBHP) system, per kg hydrogen produced, are found to be 2515 g CO₂ eq and 11.252 g SO₂-eq, respectively.

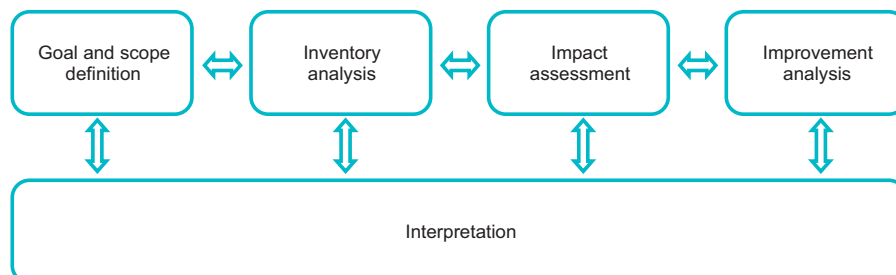


Fig. 1. Framework for life cycle assessment.

Solli, Stromman, and Herrtwich (2006) performed a comparative hybrid life cycle assessment for nuclear-assisted TWS process using the sulphur–iodine (S–I) cycle and for natural gas steam reforming with CO₂ sequestration. TWS is shown to have lower environmental impacts in terms of GWP and AP. While the GWP of natural gas steam reforming is 1.3×10^4 kg CO₂-eq, the GWP of TWS via the S–I cycle is 2.9×10^3 kg CO₂-eq for the production of 1 TJ (on the basis of higher heating value) of hydrogen. An overall advantageous option could not be determined since a weighting method was not applied.

Orhan, Dincer, and Rosen (2011) analyzed a coupling of nuclear and renewable energy sources for hydrogen production by the Cu–Cl TWS cycle, to determine the most appropriate option for the Cu–Cl cycle. An environmental impact assessment is conducted and compared with conventional hydrogen production methods using fossil fuels and other options. The results showed that CO₂ emissions for hydrogen production are negligibly small from renewables, and highest for the coal based Cu–Cl cycle.

Lubis, Dincer, and Rosen (2010) performed a preliminary LCA of a system which utilizes nuclear energy to drive the Cu–Cl TWS cycle for hydrogen production. CML 2001 impact categories are used in the LCIA and results show that the GWP of the system is 0.0025 g CO₂-eq, and that construction of nuclear and hydrogen plants are major contributors to the GWP.

4. System description and analysis

Nuclear-based hydrogen production (NBHP) via TWS system is composed of three main subsystems: fuel (uranium) processing, nuclear plant, and hydrogen plant. Mining, milling, conversion, enrichment and fuel fabrication are the main steps of the fuel (uranium) processing. The output of the fuel processing, which is fabricated uranium (UO₂), is then transported to the nuclear plant. The thermal and electrical energy output of the nuclear plant is required for the hydrogen plant, in order to produce the final output, hydrogen. In this assessment, only the construction and

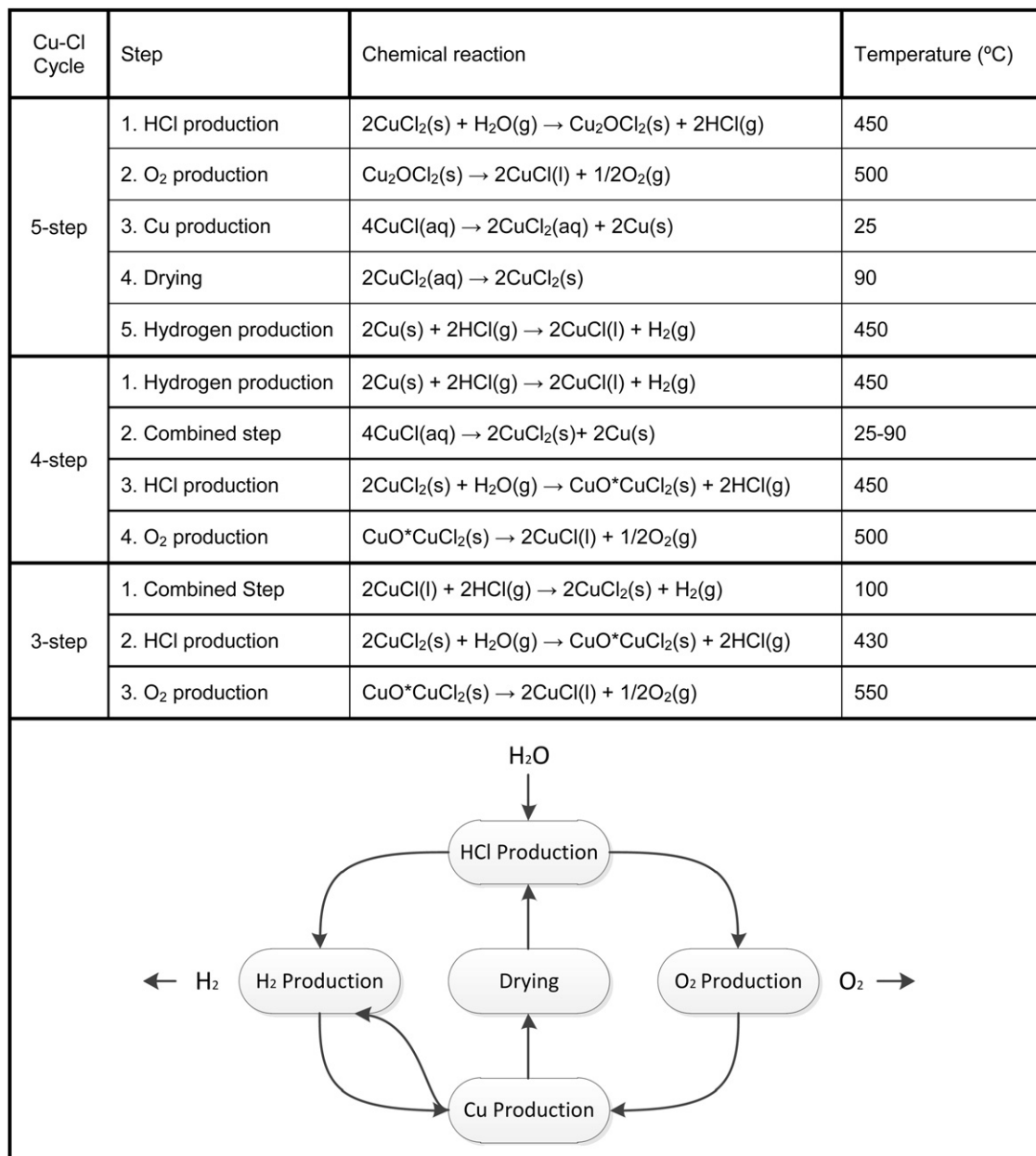


Fig. 2. Principal chemical reactions in five-, four- and three-step Cu–Cl cycles for thermochemical water decomposition and conceptual layout of the five-step cycle.

operation stages of the NBHP system are included, to be consistent with other LCA studies in the literature. Data required for the LCI phase are obtained from literature and normalized according to the method considered here.

The Cu–Cl thermochemical cycles involve a series of chemical reactions, and are characterized by the number of major chemical steps they incorporate. Two-, three-, four- and five-step Cu–Cl cycles for TWS have been defined previously in the literature. The chemical reactions used in the Cu–Cl TWS cycle, which utilizes a series of copper and chloride compounds, form a closed loop, and all chemicals are recycled without any emissions (Naterer, Gabriel, Wang, Daggupati, & Gravelins, 2008). Inputs to the Cu–Cl TWS cycle are water, thermal energy and electricity, while oxygen and hydrogen are the outputs.

4.1. Hydrogen production plant based on a Cu–Cl cycle linked with a generation IV SCWR

This study considers three main types of Cu–Cl cycles for TWS (three-, four- and five-step). A conceptual schematic of the five-step Cu–Cl cycle is shown in Fig. 2, which describes the steps, chemical reactions and corresponding temperatures in the five-, four- and three-step Cu–Cl cycles.

The five-step Cu–Cl cycle considered here consists of the following main steps: (1) HCl (g) production, (2) O₂ production, (3) Cu production, (4) drying and (5) H₂ production. The four-step cycle the 3rd and 4th steps in the five-step cycle, whereas the three-step Cu–Cl cycle combines the H₂ production step and the combined step in the four-step cycle to reduce the complexity and equipment requirements.

The total energy requirements of the five-step Cu–Cl cycle is calculated using the study of Wang, Naterer, Gabriel, Gravelins, and Daggupati (2010), in which heat requirements for each step of the five-step Cu–Cl TWS cycle are evaluated (Table 1). Table 1 shows that the total required heat input of the five-step Cu–Cl cycle is 554.7 kJ/mol H₂ and the total heat output of the cycle is 232 kJ/mol H₂. The external thermal energy requirement of the

five-step Cu–Cl cycle is 391.4 kJ/mol H₂, i.e., 195.7 MJ/kg H₂ assuming 70% heat recovery, i.e. only low grade heat is recovered. The electrical energy required for the Cu production step is 62.6 kJ/mol H₂. Also, 38 kJ/mol H₂ of work is estimated to be required for auxiliary equipment (Rosen, Naterer, Chukwu, Sadhankar, & Suppiah, 2012). Hence, the five-step Cu–Cl cycle needs 100.6 kJ/mol H₂, i.e., 50.3 MJ/kg H₂ electrical energy. Energy requirements of the three- and four-step Cu–Cl TWS cycles are calculated using the approach followed by Rosen et al. (2012). Overall, thermal energy inputs are 195.7, 161.05 and 182.74 MJ, and electrical energy inputs are 50.3, 67.15 and 67.15 MJ for the five-, four- and three-step cycles, respectively, and 9 kg of water to the hydrogen plant. 8 kg of oxygen and 1 kg hydrogen are the outputs of the Cu–Cl cycles.

The thermal energy requirements for the Cu–Cl cycles become the basis of the present analysis of the integrated system. The electrical energy needed by the hydrogen plant and other processes (e.g. heavy water production, uranium milling) is met using power output of the nuclear plant. In addition, ratios of electrical energy produced to thermal energy (process heat and waste heat) are found using the study of Pioro and Duffey (2007).

Uranium to be used in the Generation IV SCWR is processed through the following steps: mining, milling, conversion, enrichment and fuel fabrication. First, uranium ore is extracted from the environment via mining, and concentrated in the form of U₃O₈ during milling. In the conversion process, the U₃O₈ is then converted to UF₆ to be ready for enrichment, which increases the concentration of the fissile isotope U-235 to a desired level (4% for a SCWR). The chemical composition is then altered to UO₂ in the fuel production step for use in the SCWR. Table 2 presents inputs and outputs to the hydrogen plant using three Cu–Cl cycles and the Generation IV SCWR. Further details on the systems considered in this study are presented elsewhere (Ozbilen, Dincer, & Rosen, 2011a).

LCAs are conducted on three systems utilized for NBHP using three-, four- and five-step Cu–Cl TWS cycles. Data (inputs/outputs for the subsystems) from previous sections, and material and energy flows from reports in the literature (Lubis et al., 2010; Solli, 2004) are used in the analyses, which are all based on one kg

Table 1
Heat requirements of individual steps of the five-step Cu–Cl.

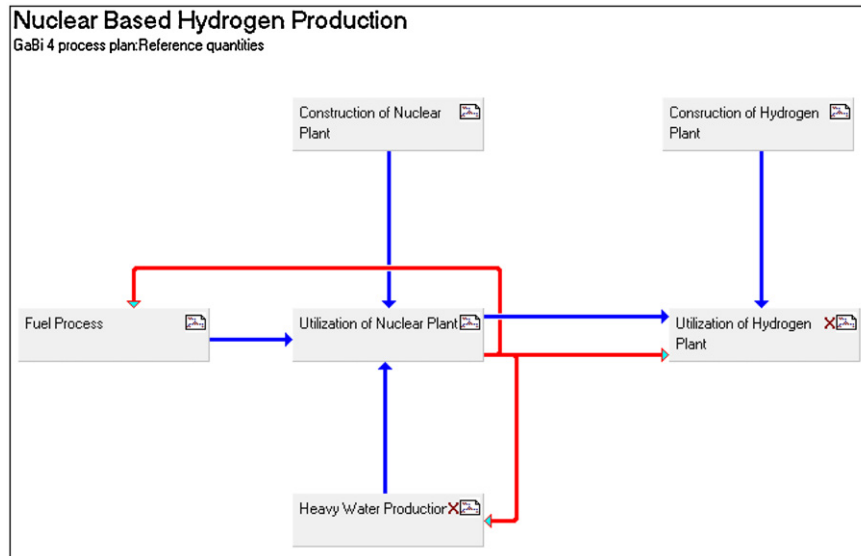
Step	Processes	Temp. (K)	Heat input (kJ/mol H ₂)		Heat output (kJ/mol H ₂)
			Total	Low-grade	
I	2Cu _(s) + 2HCl _(g) → 2CuCl _(l) + H _{2(g)}				
	Hydrogen production step				
	Vaporizing moisture from Cu _(s)	308 → 343	29.0	29.0	
	Heating Cu _(s)	298 → 723	23.4	0.1	
	Heating HCl _(g)	298 → 723	3.0	0.3	
	Heat of reaction	723			46.8
	Cooling and solidification of molten CuCl	723 → 298			80.8
	Cooling of H ₂ product	723 → 298			12.2
II	4CuCl _(aq) → 2CuCl _{2(aq)} + 2Cu _(s)				
	Electrolysis step in HCl solution	308 → 343	62.6 (electrical energy)		
III	2CuCl _{2(aq)} → 2CuCl _{2(s)}				
	Drying step				
	Vaporizing water from CuCl ₂ precipitate	308 → 343	122.0	122.0	
IV	2CuCl _{2(s)} + H ₂ O _(g) → CuO-CuCl _{2(s)} + 2HCl _(g)				
	Hydrolysis step				
	Heating CuCl _{2(s)}	298 → 648	54.2	8.5	
	Heat of reaction	648	116.6		
	Steam production H ₂ O _(l) = H ₂ O _(g)	289 → 648	57.1	3.4	
V	CuO-CuCl _{2(s)} → 2CuCl _(l) + 1/2O _{2(g)}				
	Oxygen production step				
	Heating CuO-CuCl _{2(s)}	648 → 803	20.2		
	Heat of reaction	803	129.2		
Sum		kJ/mol H ₂	554.7	163.3	232

Source: Wang et al. (2010).

Table 2

Overall inputs and outputs (per kg hydrogen produced) for hydrogen and nuclear plants using the five-, four- and three-step Cu–Cl cycle.

Plant section	Inputs	Outputs				Outputs		
		5-step	4-step	3-step		5-step	4-step	3-step
Hydrogen Production	Heat (MJ)	195.7	161.1	182.7	Hydrogen (kg)	1	1	1
	Electricity (MJ)	50.3	67.2	67.2	Oxygen (kg)	8	8	8
	Water (kg)	9	9	9				
Nuclear (SCWR)	Uranium (g)	0.404	0.333	0.377	Electricity (MJ)	313.1	257.7	292.4
					Reactor heat (MJ)	195.7	161.1	182.7
					Waste energy (MJ)	195.7	161.1	182.7

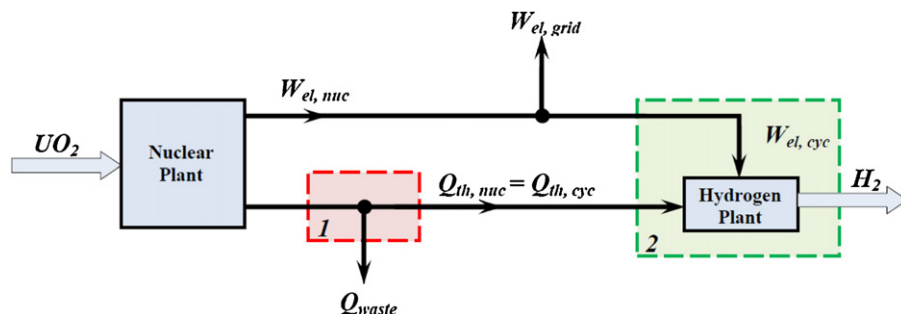
**Fig. 3.** LCA model of overall nuclear-based hydrogen production system based on thermochemical water decomposition using Cu–Cl cycles. (For interpretation of the references to color in text, the reader is referred to the web version of the article.)

hydrogen production. The overall system and all of its stages are modeled using GaBi 4 LCA software. Fig. 3 illustrates the overall system modeled in GaBi 4. First, sub-processes, such as fuel fabrication, heavy water production, etc., are modeled and then they are combined to subsystems, i.e. fuel processing, the nuclear plant and the hydrogen plant. Then, these are all linked as seen in Fig. 3 in order to analyze the environmental performance and to determine the overall environmental impact of the systems. The nuclear plant is assumed to supply the entire electrical energy requirement of each process. The red line in Fig. 3 denotes an electrical energy transfer, and is required since excess generated electricity is sent to the power grid. The processes having sub-plans are indicated by the icon on the top right corner of each of the processes. For example, fuel processing has sub-plans including mining, milling, etc. Detailed information about the GaBi 4 model of the system and its analysis is presented elsewhere (Ozbilen, 2010).

4.2. Thermal management opportunities

The aim of thermal management is to find possible ways to reduce defined environmental impacts. It is observed that fossil fuel use in the processes of the fuel cycle, which is directly proportional to nuclear reactor energy output, is the greatest contributor to almost all impact categories. Moreover, as mentioned earlier, reactor energy output is dependent on the Cu–Cl cycle thermal energy requirement. Therefore, it is determined that the four-step Cu–Cl cycle, which has the lowest thermal energy requirement of the cycles, has lower environmental effects than the three- and five-step Cu–Cl cycles.

Fig. 4 shows a simplified system diagram. UO_2 is the main input to the nuclear plant; electrical energy ($W_{el,nuc}$), waste heat (Q_{waste}) and thermal energy ($Q_{th,nuc}$) are the outputs from the system. A small portion of the electricity produced by the nuclear plant as

**Fig. 4.** Simplified nuclear-based hydrogen production system diagram.

well as the thermal energy are the inputs to the hydrogen plant, and H_2 is the ultimate output.

In this system (Fig. 4), two thermal management options, outside of the hydrogen plant and within the hydrogen plant, are considered by defining two efficiencies and shown by boxes with dashed lines (labeled 1 and 2).

Applying thermal management to the nuclear plant and to heat transfer from the nuclear plant to the hydrogen plant allows an increase in the thermal output ratio, which is defined as follows:

$$tor = \frac{Q_{th,nuc}}{Q_{th,nuc} + Q_{waste}} \quad (2)$$

Fig. 4 also shows that electrical and thermal energy are outputs of the nuclear plant. Thermal energy output comes from both waste and process heat. As previously mentioned, the hydrogen plant uses the waste and process heat from the nuclear plant. This part of thermal management deals with how much thermal energy can be obtained from the waste and process heat. The thermal output ratio directly affects not only the heat transfer to the hydrogen plant but also the mass of uranium required. Hence, the variation of environmental impacts with respect to thermal output efficiency is investigated.

As mentioned in Section 4.1, along with heat inputs (such as endothermic reactions and heat requirement before reactions), there are also heat outputs from exothermic reactions and heat exchangers in the Cu–Cl cycle (Table 1). Increasing the heat recovery fraction (hrf), and therefore heat recovery, will cause a reduction in the thermal energy requirement of the Cu–Cl cycle. There are various heat recovery options that can affect the heat recovery fraction. For example:

- selecting more efficient heat exchangers in the cycle, which have higher material qualities;
- choosing more efficient secondary fluids in the heat exchangers, which have higher heat capacity; and
- approaching stoichiometric reactions in each step.

Due to the lack of reports in the literature on applications of the previously mentioned thermal management options or experimental set-up to perform these analyses, the effects of heat recovery to the environment is investigated in this study by changing the heat recovery fraction from 0 to 1, which ultimately affects the hydrogen plant efficiency. The second box in Fig. 4 represents the second thermal management option which is related to the efficiency of the hydrogen plant. The output is the hydrogen, hence the lower heating value (LHV) of hydrogen is in the numerator and the inputs (thermal energy and electrical energy from nuclear plant) are in

the denominator. Thus, the efficiency of hydrogen plant can be calculated as follows:

$$\eta_{cyc} = \frac{LHV_{H_2}}{Q_{th,cyc} + W_{el,cyc}} = \frac{LHV_{H_2}}{Q_{endo,cyc} - hrf \times Q_{exo,cyc} + W_{el,cyc}} \quad (3)$$

where $Q_{exo,cyc}$ is the total heat released by exothermic reactions, $Q_{endo,cyc}$ is the reaction heat for endothermic reactions in the cycle, LHV_{H_2} is the lower heating value of hydrogen, which is equal to 120 MJ/kg, and $W_{el,cyc}$ is the electrical energy needed by the cycle.

As seen in Eq. (3), the efficiency of the hydrogen plant is related to energy requirements of the Cu–Cl cycle which affects environmental impacts of the NBHP system.

5. Results and discussion

The variation of environmental impacts in terms of two impact categories (AP and GWP), with the thermal output ratio of the nuclear plant and the hydrogen plant efficiency are presented. First, environmental impacts of the NBHP using the three-, four-, and five-step Cu–Cl thermochemical cycles are investigated by altering the thermal output ratio from 0.1 to 1. Second, environmental effects are calculated by varying the hydrogen plant efficiency from 0.34 to 0.65. The environmental impacts are presented for 1 kg hydrogen production.

5.1. Variation of environmental impacts with thermal output ratio

Environmental impacts associated with varying thermal output efficiency are studied in this section. Thermal output efficiency can be increased by reducing the waste heat. For example, eliminating the waste heat will result 100% thermal output efficiency. The results are presented in Figs. 5 and 6 in terms of GWP and AP and show that a reduction of waste heat significantly affects the environmental performance of NBHP system.

Fig. 5 shows the importance of thermal management for heat transfer from the nuclear plant to the hydrogen plant. An increase in the thermal output ratio from 0.1 to 1 decreases the GWP from 3.32 to 0.346 kg CO_2 -eq for the five-step Cu–Cl cycle, from 2.73 to 0.287 kg CO_2 -eq for the four-step cycle and from 3.10 to 0.324 kg CO_2 -eq per kg for the three-step cycle. It is not feasible to increase the thermal output ratio to 1 due to inevitable losses to the surroundings, but it is useful to consider this limit when assessing improvement methods.

The variation of AP with the thermal efficiency of heat transfer from the nuclear plant to the hydrogen plant is presented in Fig. 6, which further indicates the importance of thermal management.

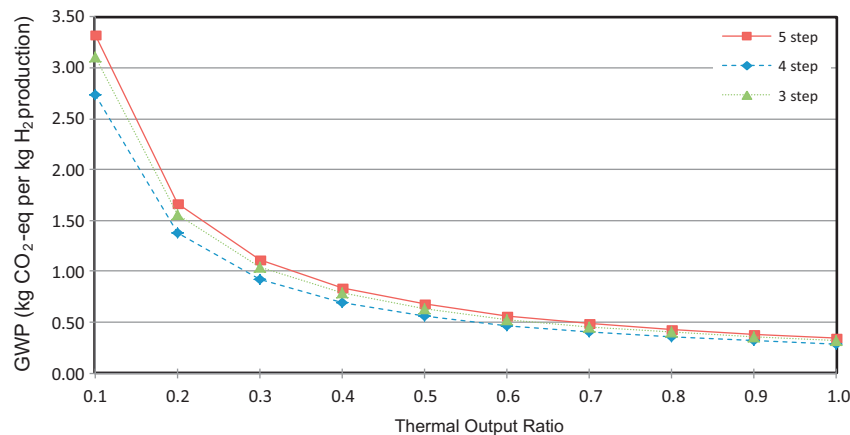


Fig. 5. Variation of GWP per kg hydrogen production with thermal output ratio for three-, four- and five-step Cu–Cl cycles for thermochemical water decomposition.

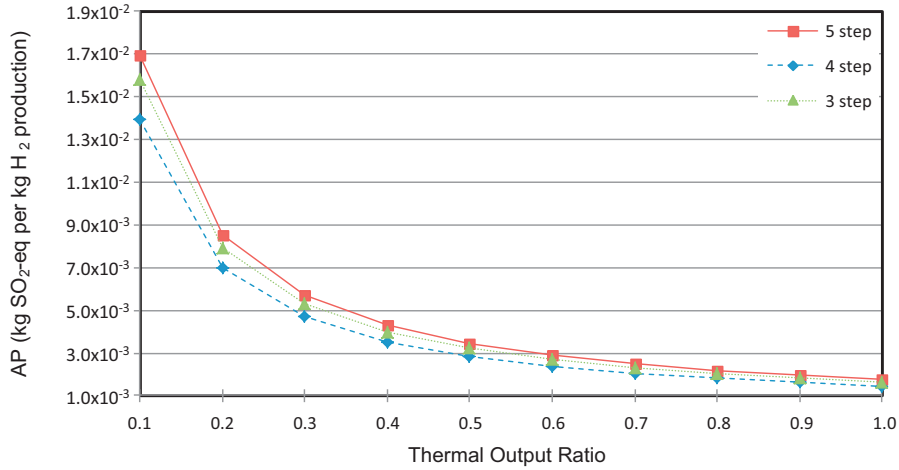


Fig. 6. Variation of AP per kg hydrogen production with thermal output ratio for three-, four- and five-step Cu-Cl cycles for thermochemical water decomposition.

For example, AP of the four-step Cu-Cl cycle is decreased from 1.40×10^{-2} kg SO₂-eq to 1.45×10^{-3} kg SO₂-eq per kg hydrogen production by increasing the thermal output ratio from 0.1 to 1. Fig. 6 also shows that the five-step Cu-Cl cycle has the highest environmental impacts for all thermal output ratio values due to it having the highest thermal energy requirement.

5.2. Variation of environmental impacts with hydrogen plant efficiency

Minimum and maximum efficiencies of the three-, four-, five-step cycles are calculated by varying the heat recovery fraction from 0, which represents no heat recovery, to 1, i.e. heat is fully recovered from exothermic reactions. The minimum and maximum efficiencies for the three-step Cu-Cl cycle can be calculated as follows:

$$\eta_{3,cyc,min} = \frac{LHV_{H_2}}{Q_{endo,cyc} + hrf \times Q_{exo,cyc} + W_{el,cyc}} = \frac{120}{284.75 - 0 \times 145.05 + 67.15} = 0.34$$

$$\eta_{3,cyc,max} = \frac{LHV_{H_2}}{Q_{endo,cyc} + hrf \times Q_{exo,cyc} + W_{el,cyc}} = \frac{120}{284.75 - 1 \times 145.05 + 67.15} = 0.58$$

Similarly, the minimum and maximum efficiencies for the four-step Cu-Cl cycle can be calculated as follows:

$$\eta_{4,cyc,min} = \frac{LHV_{H_2}}{Q_{endo,cyc} + hrf \times Q_{exo,cyc} + W_{el,cyc}} = \frac{120}{262.6 - 0 \times 145.05 + 67.15} = 0.36$$

$$\eta_{4,cyc,max} = \frac{LHV_{H_2}}{Q_{endo,cyc} + hrf \times Q_{exo,cyc} + W_{el,cyc}} = \frac{120}{262.6 - 1 \times 145.05 + 67.15} = 0.65$$

while the minimum and maximum efficiencies for the five-step Cu-Cl cycle can be calculated as follows:

$$\eta_{5,cyc,min} = \frac{LHV_{H_2}}{Q_{endo,cyc} + hrf \times Q_{exo,cyc} + W_{el,cyc}} = \frac{120}{277.35 - 0 \times 116 + 50.3} = 0.36$$

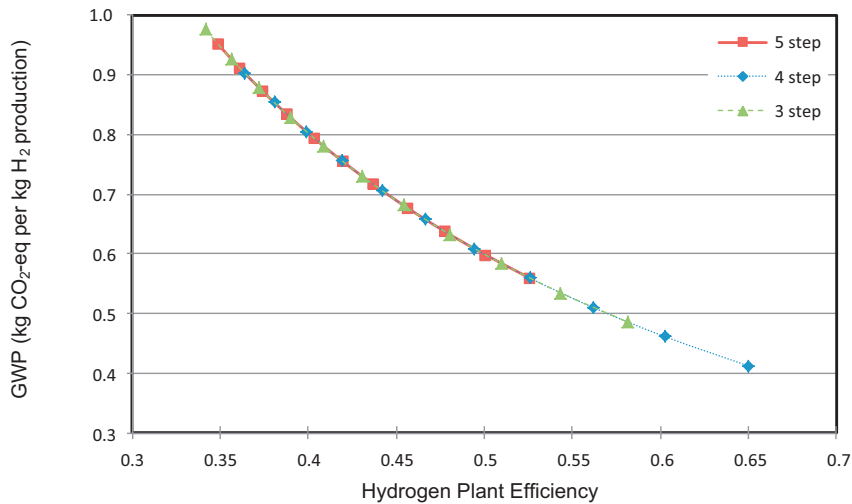


Fig. 7. Variation of GWP per kg hydrogen production with hydrogen plant efficiency for three-, four- and five-step Cu-Cl cycles for thermochemical water decomposition.

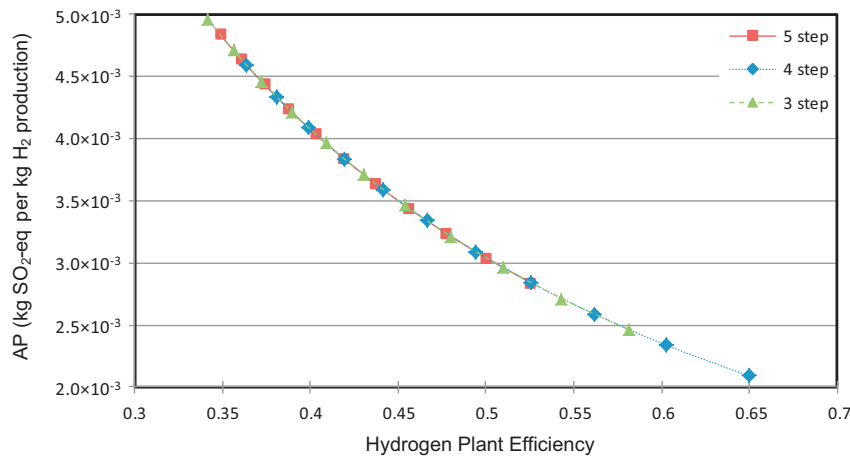


Fig. 8. Variation of AP per kg hydrogen production with hydrogen plant efficiency for three-, four- and five-step Cu–Cl cycles for thermochemical water decomposition.

$$\eta_{5, \text{cyc}, \text{max}} = \frac{LHV_{H_2}}{Q_{\text{endo}, \text{cyc}} + hrf \times Q_{\text{exo}, \text{cyc}} + W_{\text{el}, \text{cyc}}}$$

$$= \frac{120}{277.35 - 0 \times 116 + 50.3} = 0.57$$

The changes in environmental impacts by varying the Cu–Cl cycle efficiencies are also examined. The results are presented in Figs. 7 and 8 in terms of GWP and AP. The significance of heat recovery can be seen in Fig. 7, which shows that GWP can be reduced to as low as 0.4 kg CO₂-eq per kg hydrogen production, and that this value is lower than that for all the hydrogen production methods presented in the study of Ozbilen, Dincer, and Rosen (2011b). Hence, further studies should be performed to improve heat recovery within the system.

Fig. 8 shows that a heat recovery process is important for reducing AP as well. All three cycles have similar trends; however it is possible to increase the efficiency of the four-step Cu–Cl more than for the three- and four-step cycles. The lowest AP is around 2.1×10^{-3} kg SO₂-eq per kg hydrogen production for fully recovered heat in the four-step Cu–Cl cycle while the highest is 5.0×10^{-3} kg SO₂-eq per kg hydrogen production for the “no heat recovery” case of the five-step Cu–Cl cycle.

6. Conclusions

The effects of thermal management on environmental impacts of nuclear-based hydrogen production are investigated using life cycle assessment. The variations of acidification potential and global warming potential with the thermal output ratio of the nuclear plant and the hydrogen production efficiency are examined. Environmental impacts associated with NBHP using the three-, four-, and five-step Cu–Cl TWS cycles are calculated by altering the thermal output ratio from 0.1 to 1. The results show that GWP can be decreased from 3.32 to 0.346 kg CO₂-eq for the five-step Cu–Cl cycle, from 2.73 to 0.287 kg CO₂-eq for the four-step cycle and from 3.10 to 0.324 kg CO₂-eq for the three-step cycle, all per kg hydrogen production. This result demonstrates that eliminating waste heat and using it for the Cu–Cl cycle significantly affects the environmental performance of the hydrogen production system. Also, environmental impacts are investigated for various hydrogen plant efficiencies, ranging from 0.34 to 0.65. Increasing the hydrogen plant efficiency to 0.65 decreases GWP to 0.4 kg CO₂-eq and AP to 2.1×10^{-3} kg SO₂-eq. The highest efficiency and the lowest environmental impacts are for the four-step Cu–Cl cycle due to its lower heat requirements.

Acknowledgement

The authors acknowledge gratefully the financial support provided by the Ontario Research Excellence Fund.

References

- Curran, M. A. (2000). Life cycle assessment: An international experience. *Environmental Progress*, 19, 65–71.
- Davison, J., Arienti, S., Cotone, P., & Mancuso, L. (2010). Co-production of hydrogen and electricity. *International Journal of Greenhouse Gas Control*, 4, 125–130.
- Dincer, I., & Balta, M. T. (2011). Potential thermochemical and hybrid cycles for nuclear-based hydrogen production. *International Journal of Energy Research*, 35, 123–127.
- Funk, J. E. (2001). Thermochemical hydrogen production: Past and present. *International Journal of Hydrogen Energy*, 26, 185–190.
- Guinée, J. B., Gorée, M., Heijungs, R., Huppes, G., Kleijn, R., Koning, A. D., et al. (2002). *Handbook on life cycle assessment. Operational guide to the ISO standards. I: LCA in perspective. IIa: Guide. IIb: Operational annex. III: Scientific background*. Dordrecht: Kluwer Academic Publishers.
- International Energy Agency (IEA). *Technical report. Key World Energy Statistics*. (2010). http://www.iea.org/Textbase/nppdf/free/2010/key_stats_2010.pdf
- International Organization for Standardization (ISO) (1997). *ISO 14040, environmental management – life cycle assessment – principles and framework*.
- International Organization for Standardization (ISO) (1998). *ISO 14041, environmental management – life cycle assessment – goal and scope definition and inventory analysis*.
- International Organization for Standardization (ISO) (2000). *ISO 14042, environmental management – life cycle assessment – life cycle impact assessment*.
- Koornneef, J., van Keulen, T., Faaij, A., & Turkenburg, W. (2008). Life cycle assessment of a pulverized coal power plant with post-combustion capture, transport and storage of CO₂. *International Journal of Greenhouse Gas Control*, 2, 448–467.
- Lubis, L. L., Dincer, I., & Rosen, M. A. (2010). Life cycle assessment of hydrogen production using nuclear energy: An application based on thermochemical water splitting. *Journal of Energy Resources Technology*, 132, 0210041–0210046.
- Naterer, G. F., Gabriel, K., Wang, Z. L., Daggupati, V. N., & Gravelins, R. (2008). Thermochemical hydrogen production with a copper–chlorine cycle. I: Oxygen release from copper oxychloride decomposition. *International Journal of Hydrogen Energy*, 33, 5439–5450.
- Orhan, M. F., Dincer, I., & Rosen, M. A. (2011). Investigation of an integrated hydrogen production system based on nuclear and renewable energy sources: A new approach for sustainable hydrogen production via copper–chlorine thermochemical cycles. *International Journal of Energy Research*, <http://dx.doi.org/10.1002/er.1926>, published online.
- Ozbilen, A. Z. (2010). *Life cycle assessment of nuclear-based hydrogen production via thermochemical water splitting using a copper–chlorine (Cu–Cl) cycle*. Master's thesis, University of Ontario Institute of Technology.
- Ozbilen, A., Dincer, I., & Rosen, M. A. (2011a). Environmental evaluation of hydrogen production via thermochemical water splitting using the Cu–Cl cycle: A parametric study. *International Journal of Hydrogen Energy*, 36, 9514–9528.
- Ozbilen, A., Dincer, I., & Rosen, M. A. (2011b). A comparative life cycle analysis of hydrogen production via thermochemical water splitting using a Cu–Cl cycle. *International Journal of Hydrogen Energy*, 36, 11321–11327.
- Pirot, I. L., & Duffey, R. B. (2007). *Heat transfer and hydraulic resistance at supercritical pressures in power engineering applications*. New York: ASME Press.
- Rosen, M. A., Naterer, G. F., Chukwu, C. C., Sadhankar, R., & Suppiah, S. (2012). Nuclear-based hydrogen production with a thermochemical copper–chlorine

- cycle and supercritical water reactor: Equipment scale-up and process simulation. *International Journal of Energy Research*, 36, 456–465.
- Solli, C. (2004). *Fission or fossil: A comparative life cycle assessment of two different hydrogen production methods*. Master's thesis, Norwegian University of Science and Technology.
- Solli, C., Stromman, A. H., & Herrtwich, E. G. (2006). Fission or fossil: Life cycle assessment of hydrogen production. *Proceedings of the IEEE*, 94, 1785–1799.
- Urbaniec, K., Friedl, A., Huisingh, D., & Claassen, P. (2010). Hydrogen for a sustainable global economy. *Journal of Cleaner Production*, 18, S1–S3.
- Utgikar, V., & Ward, B. (2006). Life cycle assessment of ISPRA Mark 9 thermochemical cycle for nuclear hydrogen production. *Journal of Chemical Technology and Biotechnology*, 81, 1753–1759.
- Wang, Z. L., Naterer, G. F., Gabriel, K. S., Gravelsins, R., & Daggupati, V. N. (2010). Comparison of sulphur–iodine and copper–chlorine thermochemical hydrogen production cycles. *International Journal of Hydrogen Energy*, 35, 4820–4830.