LIFE CYCLE ASSESSMENT OF NUCLEAR-BASED HYDROGEN PRODUCTION USING THERMOCHEMICAL WATER DECOMPOSITION: EXTENSION OF PREVIOUS WORK AND FUTURE NEEDS

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Abstract

An extension of a previous Life Cycle Assessment (LCA) of nuclear-based hydrogen production using thermochemical water decomposition is reported. The copper-chlorine thermochemical cycle is considered, and the environmental impacts of the nuclear and thermochemical plants are assessed, while future needs are identified. Environmental impacts are investigated using CML 2001 impact categories. The nuclear fuel cycle and construction of the hydrogen plant contribute significantly to total environmental impacts. The environmental impacts for the operation of the thermochemical hydrogen production plant contribute much less. Changes in the inventory of chemicals needed in the thermochemical plant do not affect significantly the total impacts. Improvement analysis suggests the development of more sustainable processes, particularly in the nuclear plant. Other important and necessary future extensions of the research reported are also provided.

1. Introduction

In the future, hydrogen is expected to be an important energy carrier and its demand is expected to rise significantly [1, 2]. Growing concerns about the environmental impact of energy use (e.g., climate change and acid precipitation) are likely to foster increased applications of hydrogen in transportation, industry and other sectors. Hydrogen needs to be produced from an energy source (e.g., fossil fuel, uranium, sustainable energy resources like wind, solar and hydraulic). The environmental advantages of hydrogen use require the consideration of, among other factors, the environmental impacts of hydrogen production. A potentially important route to large-scale hydrogen production is thermochemical water decomposition using heat from nuclear power technologies, and one such process is the subject of an active research project being led by University of Ontario Institute of Technology.

Life cycle assessment (LCA) is a useful tool for evaluating the environmental impacts of technologies over their lifetimes, from natural resource extraction and plant construction to distribution and final product utilization. Adequate evaluation of environmental emissions and energy use throughout the overall production and utilization life cycle ("from cradle to grave") and all of its processes is critical for the proper evaluation of technologies.

The objective of this paper is to investigate using LCA the environmental impacts of nuclear-based hydrogen production. The specific production process targeted for application is based on thermochemical water decomposition. The copper-chlorine (Cu-Cl) thermochemical cycle, driven by nuclear energy, is considered. The entire life cycle of nuclear-based

thermochemical hydrogen production is considered, allowing environmental issues to be quantified and related specifically to the part of the life cycle that is responsible for them.

2. Background

2.1 LCA

LCA generally involves four main stages (Fig. 1): goal and scope definition for a study, inventory analysis, impact assessment and improvement assessment. Numerous approaches to LCA have been reported. The LCA methodology and framework of the International Organisation for Standardisation (ISO), as published in guideline ISO 14044 [3], is used here.



Figure 1. The four main stages of LCA, and the interactions between them.

2.2 LCA of hydrogen production and related technologies

Life cycle assessments have been performed of hydrogen production from a fossil fuel (natural gas) and renewable energy (solar and wind energy) [4-7]. For example, the potential of hydrogen to alleviate many environmental impacts and concerns was demonstrated by comparing the utilization of hydrogen instead of natural gas in a combustion engine and an electrical power plant.

During the last decade, little effort has been directed towards LCAs of hydrogen production using nuclear energy and related technologies. Utgikar and Thiesen [8] conducted an LCA of hydrogen production via electrolysis driven by electricity from a nuclear power facility. In another study, Solli et al. [9] applied LCA to two hydrogen production processes, comparing the environmental impacts of hydrogen production from nuclear energy to that from natural gas.

Recently, Lubis et al. [10] performed a preliminary LCA nuclear-based hydrogen production using thermochemical water splitting. It was found that the nuclear fuel cycle contributes significantly to the environmental impacts of the overall system. This paper is an extension of the work reported in that study.

3. Thermochemical water decomposition process considered for hydrogen production

In thermochemical water decomposition, water and heat are supplied to the process and oxygen and hydrogen extracted, while numerous chemicals are recycled and used in the same reactions in subsequent cycles. The overall net chemical reaction is:

$$H_2O \to H_2 + \frac{1}{2}O_2$$
 (1)

The copper-chlorine (Cu-Cl) thermochemical cycle for hydrogen production from water, one of several thermochemical cycles proposed [11], is considered here. Heat from the nuclear

(4)

reactor is supplied to various chemical reactions in the thermochemical hydrogen production plant at various temperatures. The thermochemical process involves thermally assisted chemical reactions that, when summed, yield the simple net reaction of water decomposition into hydrogen and oxygen. Argonne National Laboratory has carried out initial research on the Cu-Cl thermochemical cycle for producing hydrogen [12]. The main reactions in the process and their respective required temperatures are shown below [13]:

$2 \operatorname{CuCl} + 2 \operatorname{HCl} (a) \to \operatorname{H}_2 (g) + 2 \operatorname{CuCl}_2$	(~100°C)	(2)

$2 \operatorname{CuCl} 2 + \operatorname{H}_2 O(\mathfrak{g}) \rightarrow \operatorname{Cu}_2 O(\mathfrak{Cl}_2 + 2 \operatorname{HCl}(\mathfrak{g}))$	$(\sim 400^{\circ}C)$	(3)
$2 CuCl2 + H_2O(g) + Cu_2OCl2 + 2 HCl(g)$	(100 C)	(\mathbf{J})

Cu₂OCl₂ → 2CuCl (l) + $\frac{1}{2}$ O₂ (g) (~400-550°C)

The system considered incorporates an advanced nuclear power plant combined with a hydrogen production plant based on the Cu-Cl thermochemical cycle. The advanced nuclear power plant provides thermal energy for driving the chemical reactions listed previously. The nuclear reactor considered is the Super-Critical Water-Cooled Reactor (SCWR), which is one of the Generation IV advanced reactors under investigation by many countries [14]. The SCWR is proposed as a Generation IV nuclear reactor for Canada. The Cu-Cl thermochemical water decomposition cycle integrates well with Canada's potential future nuclear reactors, since the temperatures at which it requires heat are adaptable to those which can be provided by the SCWR.

4. Previous LCA of nuclear-based thermochemical hydrogen production

A preliminary LCA by the authors of nuclear-based hydrogen production via the copper-chlorine (Cu-Cl) thermochemical water decomposition cycle (Fig. 2) has been reported [10]. The overall thermal output of the nuclear plant is assumed dedicated to producing hydrogen and the nuclear plant is rated at 2060 MWth [15]. The production capacity of the thermochemical plant is taken to be 3800 kg H_2/h [10] with 30 years of operational life. That previous work is summarized and extended in this section, considering each of the four main stages of an LCA separately.

4.1 Goal and scope definition

An LCA of the hydrogen production system in Fig. 2 is conducted to estimate its environmental impacts. The emissions from the overall system are considered the sum of the emissions from the advanced nuclear power plant and the thermochemical hydrogen production plant. The following CML 2001 impact categories [16] are used:

- Abiotic resource depletion potential (ADP) (in g extracted element), which is a measure of the extraction of non-renewable raw materials.
- Global warming potential (GWP) (in g CO₂-equivalent), which is caused by increases in CO₂ in the earth's atmosphere.
- Ozone depletion potential (ODP) (in g CFC-equivalent), which leads to an increase in ultraviolet radiation reaching the earth's surface.
- Eutrophication potential (EP) (in kg phosphate-equivalent), caused by over-fertilisation or nutrition enrichment.
- Acidification potential (AP) (in g SO2-equivalent), which can change acidity of soil and water.

- Photochemical ozone creation potential (POCP) (in kg ethene-equivalent), due to volatile organic compounds in the atmosphere.
- Radioactive radiation (RAD) (in disability-adjusted life years, DALY), which measures the emission and propagation of energy in the form of rays or waves.



Figure 2. System description for the LCA.

4.2 Inventory analysis

The inventory analysis estimates the environmental impacts specified in the previous subsection. The overall emissions are the sum of the emissions from the nuclear power and thermochemical hydrogen production plants.

For the nuclear plant, environmental emissions are mainly from the nuclear fuel cycle and plant construction and installation. Where exact information and data are not available, the quantities and types of materials and energy used are assumed to be similar to those for current nuclear plants. The Ministry of Commerce, Industry, and Energy of Korea has reported emissions of a nuclear fuel cycle covering the activities from mining of the ore to spent fuel disposal [17], and this information was used to estimate emissions from the nuclear plant considered here.

Energy use associated with heavy water production is based on records from the Bruce heavy water plants [18]. The SCWR reactor is somewhat different from other reactor technologies in reliance on heavy water to achieve a nuclear reaction with uranium fuel. The amount of uranium fuel needed M to produce 1 MWth heat is calculated as

$$M = \frac{Q}{B_d}$$
(5)

where Q is heat produced and B_d is the discharge burn-up, assumed to be 480 MWh/kg (U) [15].

Regarding the physical plant construction, data is based on reported data by Vattenfall [19], who estimate the quantities and types of materials used in nuclear plant construction as well as operating emissions. Data are modified where necessary. Process inputs are shown in Table 1. Emissions for each of the steps described in Table 1 are also considered, but are not listed because they are numerous.

Table 1. Inputs for the inventory analysis for the process in the nuclear plant considered

Mining 1 kg uranium ore	1
Economic inflows	
Electricity	36.8 MJ
Equipment fuel	8.55 MJ
Economic outflows	
Uranium ore (underground mine) U	1 kg
Environmental resources	
Land	0.156 m^2
Water	0.00213 m ³
Milling 1 kg uranium	
Economic inflows	
Electricity	41 MJ
Heating fuel	83.8 MJ
Economic outflows	
Uranium in milling	1 kg
Environmental resources	
Land	0.75 m ²
Water	0.456 m^3
Conversion of 1 kg uranium	
Economic inflows	
Electricity	31.7 MJ
Coal	115,000 MJ
Natural gas	166 MJ
Economic outflows	
Uranium natural in UF6	1 kg
Environmental resources	
Water	0.661 m^3
Economic inflows	
Electricity	6.61x10 ⁻⁵ MJ
Coal	94.8 MJ
Gasoline	1.68 MJ
Economic outflows	
Enriched uranium	1 kg
Environmental resources	
Water	1.94 m^3
Fabrication of fuel	•
Economic inflows	
Electricity	38.1 MJ
Coal	108 MJ

Natural gas	28.8 MJ
Economic outflows	
Fabrication uranium	1 kg
Environmental resources	
Water	0.123 m ³
Production of 1 tonne heavy water	۴
Economic inflows	
Electricity	6600 MWh
Economic outflows	
Heavy water	1000 kg
Environmental resources	
-	-
Construction of nuclear plant	
Economic inflows	
Aluminium ingots	29,040 kg
Concrete	5.227 x 10 ⁷ kg
Copper	40,650 kg
Lead	65,820 kg
Polyvinylchloride (PVC)	225,300 kg
Spruce	4.956 x 10 ⁶ kg
Steel	$1.328 \times 10^7 \text{ kg}$
Titanium	19,360 kg
Economic outflows	
Nuclear power plant	1 plant
Environmental resources	
-	-
Economic inflows	
Aluminium ingots	0.00434 kg
Copper	0.036 kg
Lead	0.00983 kg
Polyvinylchloride (PVC)	0.036 kg
Steel	0.132 kg
Titanium	0.000289
Heavy water	22,000 kg
Economic outflows	
Nuclear heat	1 TJ
Environmental resources	
-	-

For the thermochemical hydrogen production plant, environmental impacts are estimated based on energy use, use of raw materials (e.g., water), and plant fabrication and installation. The energy required to operate the thermochemical plant is provided by the nuclear power plant, and is accounted for in its assessment. All relevant chemical species are seen from Eqs. (1-3) to be obtainable from CuCl and HCl. The inventory of this chemical and water as a raw material are needed to estimate emissions. Lubis et al. [10] have estimated the inventories for the chemicals and raw material needed for a hydrogen plant to produce 3800 kg hydrogen using the Cu-Cl thermochemical cycle: 133.2 tonnes of CuCl, 384 tonnes of HCl and 40 tonnes of H₂O.

The environmental emissions associated with the fabrication and installation of the thermochemical plant depend on the quantities and types of materials used. General Atomic has estimated this information [20] for hydrogen production with the sulphur-iodine thermochemical cycle. This information is used here, changing the chemical species to copper chloride (CuCl) and applying relevant assumptions. Process inputs for the thermochemical plant are listed in Table 2. Corresponding emissions for the steps in Table 2 are also considered but are too numerous to list.

Construction of hydrogen plant			
Economic inflows			
Aluminium ingots	22,260 kg		
Concrete	40,060,000 kg		
Copper	311,600 kg		
Lead	50,450 kg		
Polyvinylchloride (PVC)	172,700 kg		
Spruce	3,799,000 kg		
Steel	10,180,000 kg		
Titanium	14,840 kg		
Economic outflows			
Hydrogen production plant	1 plant		
Environmental resources			
_	-		
Production of 3800 kg hydrogen			
Economic inflows			
Copper chloride	133,200 kg		
Hydrogen chloride	384,000 kg		
Water (raw materials)	40,000 kg		
Aluminium ingots	0.002362 kg		
Copper	0.019615 kg		
Lead	0.005346 kg		
Polyvinylchloride (PVC)	0.018346 kg		
Steel	0.071923 kg		

Table 2. Inputs for the inventory analysis for the processes in the thermochemical hydrogen plant
considered.

Titanium	0.000315 kg
Economic outflows	
Hydrogen	3800 kg
Environmental resources	
_	

4.3 Impact assessment

For the CML 2001 impact categories and the data in Tables 1 and 2, the GaBi lifecycle database (created with industry) [16] is used to determine environmental impact values (see Table 3).

Table 3. Values for several environmental impact categories for nuclear-based hydrogen

production.	
Environmental impact (EI) category	
ADP (g extracted element)	1.45 x10 ⁻⁰⁷
AP (g SO ₂ -eq.)	$1.48 \text{ x} 10^{-04}$
EP (kg phosphate-eq.)	$1.04 \text{ x} 10^{-04}$
GWP (g CO ₂ -eq.)	$2.51 \text{ x} 10^{-03}$
ODP (g CFC-eq.)	$7.16 \text{ x} 10^{-07}$
POCP (kg ethene-eq.)	$1.41 \text{ x} 10^{-04}$
RAD (DALY)	1.98×10^{-05}

4.4 Improvement assessment

Several modifications are possible to reduce the environmental impacts associated with the nuclear-based hydrogen production process. These are examined in the following manner:

- The effects of changing the quantity of materials for the thermochemical plant are estimated.
- The environmental impacts associated with the nuclear power plant are investigated assuming actual emissions are 1) 50% higher due to the increased plant complexity, and 2) 50% lower than those considered here due to higher burn up and more efficient utilization of nuclear fuel.

Environmental impact category	Base case	Materials need doubled	Chemical inventory doubled	Nuclear plant emissions doubled	Nuclear plant emissions halved
ADP (g extracted element)	1.45x10 ⁻⁰⁷	1.45x10 ⁻⁰⁷	1.45x10 ⁻⁰⁷	2.89x10 ⁻⁰⁷	7.23x10 ⁻⁰⁸
AP (g SO ₂ -eq.)	1.48x10 ⁻⁰⁴	2.05x10 ⁻⁰⁴	1.50x10 ⁻⁰⁴	2.39x10 ⁻⁰⁴	1.03x10 ⁻⁰⁴
EP (kg phosphate-eq.)	1.04x10 ⁻⁰⁴	1.44x10 ⁻⁰⁴	1.05x10 ⁻⁰⁴	1.67x10 ⁻⁰⁴	7.24x10 ⁻⁰⁵
GWP (g CO ₂ -eq.)	2.51x10 ⁻⁰³	3.58x10 ⁻⁰³	2.52×10^{-03}	3.95×10^{-03}	1.79×10^{-03}
ODP (g CFC-eq.)	7.16x10 ⁻⁰⁷	1.02x10 ⁻⁰⁶	7.17x10 ⁻⁰⁷	1.13x10 ⁻⁰⁶	5.11x10 ⁻⁰⁷
POCP (kg ethene-eq.)	1.41x10 ⁻⁰⁴	2.00x10 ⁻⁰⁴	1.42×10^{-04}	2.21x10 ⁻⁰⁴	1.01x10 ⁻⁰⁴
RAD (DALY)	1.98x10 ⁻⁰⁵	2.25x10 ⁻⁰⁵	1.98x10 ⁻⁰⁵	3.69x10 ⁻⁰⁵	1.13x10 ⁻⁰⁵

Table 4. Sensitivity analysis of environmental impacts.

A sensitivity analysis of the environmental impacts summarizes the improvement assessment (Table 4). The base case is the sum of emissions from an advanced nuclear power plant and a thermochemical hydrogen production plant, seen in Table 3. Some key observations in Table 4 follow. Global warming potential and acidification potential, major environmental concerns, are significant. For global warming potential, the system emission is 0.00251 g CO₂-eq. over the life of the plant. This quantity is mainly attributable to the nuclear plant and construction of the hydrogen plant, which contribute 57% and 42% respectively of the total emission. For acidification potential, the system emission is 0.000148 g SO₂-eq. For this quantity, construction of nuclear plant, construction of hydrogen plant and the nuclear fuel cycle contribute 51%, 38% and 9%, respectively. Also, the environmental impacts for the chemical inventory of the total emissions. Changes in the inventory of chemicals needed in the thermochemical plant do not affect significantly the total impacts. In contrast, reducing the emissions from nuclear plant can contribute significantly in reducing the total emissions. By halving nuclear plant emissions, global warming potential and acidification potential are reduced by 28% and 30%, respectively.

5. Future work and needs

The analysis presented here is in many ways preliminary, and should be further extended so as to improve its accuracy and meaningfulness. In part the preliminary nature of the work reported is due to the fact that the technology is new and reliable data is not available for many parameters. Consequently, numerous estimations and approximations are used and the accuracy of these, and any subsequent LCAs, will be improved as research on thermochemical hydrogen production continues. More broadly, there are several areas in which needs exist for research, and these are discussed in the following subsections.

5.1 Impact of LCA on energy use and environmental impact

In 2003 primary worldwide energy consumption was about 10.7 Gtoe as reported by the International Energy Agency [21]. Eighty-one percent of this energy has been transformed into heat, electricity or movement, and has utilized fossil fuel combustion, which yields CO_2 emissions to the atmosphere [21]. If current global trends showing increasing primary energy consumption and CO_2 emissions are not altered, the risk of global climate change will increase. Decisions must be made on whether societies want to rely on current patterns of energy use or switch to more sustainable and clean energy paths. The LCA reported here can help inform such debates, and facilitate comprehensive understanding of the benefits of nuclear-based hydrogen production based on thermochemical water decomposition.

5.2 Extension of LCA to include exergy

Numerous authors have extended LCA by considering exergy instead of or with energy [22-25]. Such an extension of LCA is referred to as exergetic life cycle assessment. As exergy is the work theoretically obtainable from a flow or system, exergy accounts for both energy quantity and quality and thus more precisely characterizes the efficiency of fossil and mineral resource consumption [26-27].

Exergy analysis is a method that uses the conservation of mass and conservation of energy principles together with the second law of thermodynamics for the analysis, design and improvement of energy and other systems. The exergy method is a useful tool for furthering the goal of more efficient energy-resource use, for it enables the locations, types and magnitudes of wastes and losses to be identified and meaningful efficiencies to be determined.

Exergetic LCA has been described and applied to several hydrogen production technologies recently [28, 29]. Specifically, an exergetic life cycle assessment is presented of four technologies (two using fossil fuels and two renewable energy forms) for producing gasoline and hydrogen and their use in internal combustion (gasoline) or fuel cell (hydrogen) vehicles. In that study, life cycle exergy efficiencies are considered, as are such other factors as capital investment efficiency and environmental impact.

An exergetic life cycle assessment could be developed and used to complement the present LCA of nuclear-based hydrogen production via thermochemical water decomposition, by providing additional insights into its performance and environmental impact.

5.3 Comparison with other technologies

The results of the LCA reported here need to be compared to and contrasted with LCAs of other technologies that may compete with nuclear-based hydrogen production via thermochemical water decomposition, so that a comprehensive understanding of the benefits and drawbacks of each can be ascertained. Then, reasoned decisions can be made on what technologies will be developed and implemented, and what potential benefits can be accrued.

5.4 Improved processes for thermochemical hydrogen production

The improvement analysis part of the LCA reported here suggests the development of process improvements for both the nuclear and thermochemical technologies involved in nuclear-based hydrogen production by thermochemical water decomposition. In particular, the LCA identifies the need for more sustainable processes, particularly in the nuclear plant and construction phase of the thermochemical hydrogen production plant.

5. Conclusions

This LCA of nuclear-based hydrogen production via thermochemical water decomposition using the copper-chlorine thermochemical cycle has estimated approximate environmental parameters and identified future needs. The LCA indicates that most of the environmental impacts are associated with the nuclear plant and construction of hydrogen plant. In fact, over 99 percent of the global warming potential is released from the nuclear plant and the construction of the hydrogen plant, while 98 percent of the total acidification potential is contributed during the deployment of the nuclear fuel cycle and construction of nuclear and hydrogen plants. This LCA highlights the fact that the nuclear plant and construction of the hydrogen plant contribute significantly to the environmental impacts of the overall system. Improvement analysis suggests the development of more sustainable processes in the nuclear plant and construction of hydrogen production. Other important future extensions of the research reported are also identified.

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