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Development of net energy ratio and emission factor for biohydrogen production pathways

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ABSTRACT

This study investigates the energy and environmental aspects of producing biohydrogen for bitumen upgrading from a life cycle perspective. Three technologies are studied for biohydrogen production; these include the Battelle Columbus Laboratory (BCL) gasifier, the Gas Technology Institute (GTI) gasifier, and fast pyrolysis. Three different biomass feedstocks are considered including forest residue (FR), whole forest (WF), and agricultural residue (AR). The fast pyrolysis pathway includes two cases: truck transport of bio-oil and pipeline transport of bio-oil. The net energy ratios (NERs) for nine biohydrogen pathways lie in the range of 1.3–9.3. The maximum NER (9.3) is for the FR-based pathway using GTI technology. The GHG emissions lie in the range of 1.20–8.1 kg CO_2 eq/kg H_2. The lowest limit corresponds to the FR-based biohydrogen production pathway using GTI technology. This study also analyzes the intensities for acid rain precursor and ground level ozone precursor.

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1. Introduction

With the focus today on the development of renewable fuels and chemicals, there is a lot of interest in the utilization of biomass feedstock for producing these fuels and chemicals. Lignocellulosic biomass, including agricultural residue (i.e. straw, corn stover), forest residue (branches and tops of the trees), whole tree, and energy crops can be used to produce a range of fuels and chemicals. One of the important fuels which can be produced from biomass is hydrogen, also called biohydrogen (Sarkar and Kumar, 2009). There are several studies available in the literature on producing biohydrogen from lignocellulosic biomass (Spath et al., 2005; Larson et al., 2005; Sarkar and Kumar, 2010a,b). These are discussed later in this paper.

The Western Canadian sedimentary basins of Alberta hold the world's largest natural reservoir of bitumen. This province is experiencing an enormous growth in the oil sands industry. Oil sands are used to produce bitumen which is further modified to make petroleum products. Currently, the fossil fuel industries of Alberta (mainly petroleum refining and upgrading) of Alberta account for 17% of the overall greenhouse gas (GHG) emissions from the province; this is the second largest share after the power generation sector (55%). Note that, GHG emissions from fossil fuel industries of Alberta correspond to about 6% of Canada's total GHG emissions (Environment Canada, 2009). The cumulative GHG emissions from the oil sands industry in Alberta increased from 16.8 to 37.2 mega tonnes of CO_{2eq} between 1990 and 2008 (Droitsch et al., 2010). Based on the projected growth of the oil sands industry in Alberta, by 2020 the emissions will be three times higher than the level for 2008 (Droitsch et al., 2010). The bitumen extracted from oil sand needs to be upgraded to synthetic crude oil. In 2009, crude bitumen production in Alberta totaled 1.5 million barrels/day, which is likely to increase to 3.2 million barrels/day by 2019 (ERCB, 2010). To upgrade each barrel of bitumen, 3–5 kg of hydrogen is essential (Ordorica-Garcia et al., 2007); therefore, the hydrogen requirement for the bitumen-upgrading industry of Alberta is enormous. Natural gas and coal are the primary sources of the hydrogen produced. Since these fossil fuels have a large carbon footprint, the production of hydrogen is solely responsible for 28% of the total GHG emissions from the oil sands industry in Alberta (Ordorica-Garcia et al., 2007). By 2030, the demand for hydrogen for bitumen upgrading is expected to increase to six times that for the base year (2003); therefore, it is necessary to mitigate the overall GHG emissions from the oil sands industry. This can be done by replacing fossil fuels with hydrogen produced from renewable biomass sources. Note that, using biohydrogen in bitumen upgrading is a regional interest of Alberta. This study assesses the biohydrogen production pathways for their possible application in a bitumen-upgrading plant. In addition to its use in bitumen upgrading, biohydrogen can be used in the transportation sector and the food, petrochemical, and manufacturing industries. The results of this study could be used in other jurisdictions for application in the areas mentioned above.

Alberta has a large amount of forest residue (3.29 million dry tonnes per year) (Sarkar and Kumar, 2010a) and agricultural

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residue (6.8, 6.3, and 0.72 million tonnes of wheat, barley, and oat straw per year, respectively) (Sultana et al., 2010) that can be allocated entirely to hydrogen production. Forest harvest residue (FR) refers to the limbs, tops and branches left behind by logging operations. In Alberta more than 90% of all logging operations involve cutting the tree in the stand, skidding it to the roadside, and delimbing tops and branches from the tree on the roadside. The stem is used or it is chipped and transported to a plant. The delimbed tops and branches are then piled up and burnt to prevent forest fires. This residue is a large potential source of biohydrogen (Sarkar and Kumar, 2010a). Agricultural residues (AR) are mainly wheat and barley straw. Most are left in the field to rot and this releases carbon back into the atmosphere. Whole forest (WF) biomass, i.e. whole tree, is used in Alberta for pulp and lumber. As the demand for paper decreases, pulp mills close. In Alberta this provides an opportunity to integrate the forest industry with the hydrocarbon industry. Utilizing different forest resources for purposes other than pulp and lumber has been discussed in early studies by the authors (Kumar et al., 2003; Sarkar and Kumar, 2009, 2010a). These biomass resources can be used to produce biohydrogen.

The different biomass sources can be directly gasified to produce hydrogen (Sarkar and Kumar, 2010a). Large scale production of hydrogen from biomass using the Battelle Columbus Laboratory (BCL) gasifier and the Gas Technology Institute (GTI) gasifier are two key technologies which have been demonstrated (Spath et al., 2005; Larson et al., 2005) and studied in detail. The techno-economic assessment of employing these technologies to produce biohydrogen in Alberta through direct gasification of WF, FR and AR employing these technologies was studied earlier (Sarkar and Kumar, 2009, 2010a). Biomass is a dispersed resource that has low bulk and energy density, resulting in high transportation cost. To increase energy density, biomass for biohydrogen production can be converted into bio-oil by fast pyrolysis. This bio-oil can later be gasified and steam-reformed to produce biohydrogen. The techno-economical feasibility of this pathway of hydrogen production for Alberta was also studied earlier by Sarkar and Kumar (2010b). They found that, even at the economic optimum plant size, the cost of producing hydrogen from biomass is significantly higher than the cost of producing it from natural gas/coal. Biohydrogen could be competitive with natural-gas/coal-based hydrogen with carbon credits. Hence, GHG mitigation provides key motivation for the development of biomass-based hydrogen.

GHG is not the only detrimental effect on the environment due to producing hydrogen from coal and natural gas; acid rain precursor (ARP) and ground level ozone precursor (GOP) are also significant environmental concerns (Spath and Mann, 2001; Koroneos et al., 2004, 2005). It is very important to estimate the environmental benefits achievable if fossil fuels are replaced with biomass for hydrogen production. The most appropriate and widely-accepted methodology for environmental-impact accounting is life cycle assessment (LCA). LCA evaluates the environmental impact from any product or system throughout its life cycle, starting with raw material production and acquisition and ending with disposal. Like environmental impact analysis, energy output–input ratio i.e. net energy ratio (NER) is also critical. Evaluating the life cycle NER for renewable systems helps with understanding the effectiveness of a particular system compared to other renewable and fossil-fuel-based systems.

The objective of this study is to quantify environmental impact in terms of emissions and NERs for different biohydrogen production pathways. This study includes biohydrogen production from three biomass sources: FR, AR and WF biomass. Three different technologies for producing biohydrogen are analyzed: BCL, GTI, and pyrolysis process. These technologies are described in subsequent sections. Nine biohydrogen production pathways are compared from a life-cycle energy-and-emissions perspective. The application targeted by this biohydrogen is bitumen upgrading. The environmental stressors considered in this study are: GHG emissions (CO_{2eq}), ARP (SO_{2eq}) and GOP (NO_x + VOC). There are few studies which do comparative analyses of different biomass feedstock conversion pathways for hydrogen production. There are some LCA studies available for producing hydrogen from different renewable resources (Koroneos et al., 2004, 2008), but none of these studies investigate different biomass conversion technologies for producing hydrogen from different biomass feedstocks. This research work performs LCA for hydrogen production using the optimum plant size for each conversion pathway that was determined earlier by the authors (Sarkar and Kumar, 2009, 2010a,b).

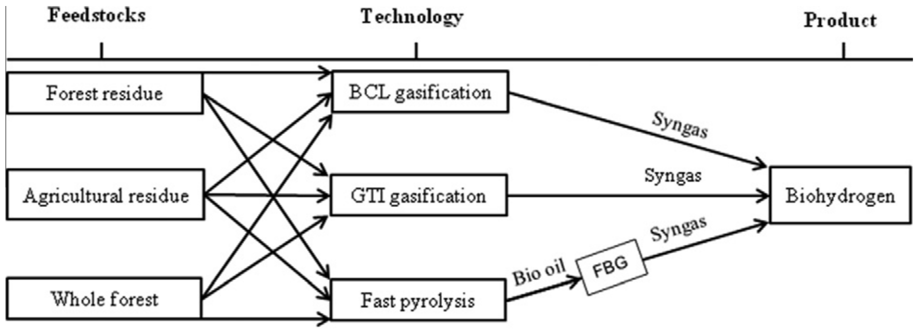
1.1. Biohydrogen production pathways

Nine different biomass conversion pathways have been considered in this study. Fig. 1 shows the different conversion pathways. Biomass is gasified at approximately atmospheric pressure (~0.16 MPa) in a BCL gasifier (Sarkar and Kumar, 2010a). In contrast, the GTI gasifier operates at a higher pressure (~3.45 MPa) to gasify the biomass (Sarkar and Kumar, 2010a). Syngas produced from biomass is purified and then undergoes a water–gas shift reaction that produces hydrogen. Details of the two biomass gasification technologies and the production of biohydrogen using these technologies were given in earlier studies (Sarkar and Kumar, 2010a; Spath et al., 2005; Larson et al., 2005). During fast pyrolysis, biomass is rapidly heated in the absence of air, it vaporizes and the organic vapors are rapidly quenched so they condense to a dark viscous liquid called bio-oil (Bridgwater and Peacocke, 2000; Sarkar and Kumar, 2010b). This bio-oil is gasified in a fluidized bed reactor (FBR) to produce syngas which is then steam reformed in the presence of noble catalysts to produce biohydrogen. Details on this pathway for the production of biohydrogen are given in Sarkar and Kumar (2010b).

This research investigates the life cycle NERs and the environmental impact of all the pathways (Fig. 1) at their respective optimum plant size. It is important to note that, optimum plant sizes are specific to location, feedstock and technology. The optimum size for each biohydrogen production plant has been derived from earlier studies by the authors for the different feedstocks available in this region. The optimum biohydrogen plant capacity for BCL pathways is 3000 dry tonnes per day (dtpd) for all three feedstocks (AR, WF, and FR) for Alberta (Sarkar and Kumar, 2009, 2010a). The capacity of each BCL gasifier unit is assumed to be 1000 dtpd (Sarkar and Kumar, 2009, 2010a; Spath et al., 2005). Optimum plant capacity for GTI pathways are 3000, 4000, and 3000 dtpd for AR, WF, and FR feedstock, respectively (Sarkar and Kumar, 2010a). The capacity of each GTI gasifier unit is assumed to be 2000 dtpd (Larson et al., 2005; Sarkar and Kumar, 2010a). Optimum plant capacity for the pyrolysis pathways is 2000 dtpd for all three feedstocks (Sarkar and Kumar, 2010b). The capacity of each pyrolysis reactor is assumed to be 1000 dtpd (Sarkar and Kumar, 2010b). The methodology of this study is further described in the following sections.

2. Methodology

Each of the biohydrogen production pathways is analyzed as a combination of several unit operations. Material, equipment, and fuel-embodied energy and emissions factors are determined for each of the unit operations involved in a conversion pathway over its life cycle. The energy and emissions associated with each of the unit operations are investigated in detail. Their impact is normalized corresponding to a common reference unit, which is called



FBG: Fluidized bed gasification, BCL: Battelle Columbus Laboratory, GTI: Gas Technology Institute

Fig. 1. Biomass conversion pathways to biohydrogen.

the functional unit (FU). FUs are crucial to any LCA study; they ensure the obtained LCA results are comparable. The FU chosen for this study is 1 kg H_2 upon delivery to a bitumen-upgrading plant. This study considers all the system's energy input to be derived for primary fossil fuel. This study evaluates the NERs for all biohydrogen production pathways, a crucial ratio for the assessment of renewable systems. The NERs for the pathways are calculated using Eq. (1).

$$\mathrm{NER} = \sum E_{\mathrm{out}} / \sum E_{\mathrm{in}} \tag{1}$$

where, $\sum E_{\mathrm{in}}$ = life cycle non-renewable primary energy input corresponding to the FU of a pathway, and $\sum E_{\mathrm{out}}$ = energy available from the FU equivalent biohydrogen produced from the pathway. Note that, this study is based on the higher heating value (HHV) for fuels. Three environmental stressors i.e. net GHG emissions, ARP, and GOP for a particular conversion pathway are calculated using Eq. (2).

$$\text{Net emission} = \sum \varepsilon_{\mathrm{out}} \tag{2}$$

where, $\sum \varepsilon_{\mathrm{out}}$ = Life cycle emissions corresponding to the FU of a pathway within the defined system boundary (Fig. 2). GHG stressors are considered to be mainly carbon dioxide (CO_2), carbon monoxide (CO), methane (CH_4), and nitrous oxide (N_2O). GHGs contribute to global warming. The global warming potential (CO_{2eq}) for these gases are assumed to be 1, 3, 21, and 310 respectively. Note that, these factors are based on a 100 year time horizon. In contrast, sulfur dioxide (SO_2) and nitrogen oxide (NO_x) are assumed to be the main ARPs. These compounds are responsible for acidification i.e. proton expulsion in the earthbound/aquatic ecosystem. With regard to ARP intensity, the weighting factor (SO_{2eq}) is assumed to be 1 and 0.7 for SO_2 and NO_x, respectively. NO_x and volatile organic compounds (VOC) are the main GOPs. The weighting factor (NO_x + VOC) is assumed to be 1 for both these compounds. Ground level ozone is a secondary form of pollutant which occurs due to chemical reaction between NO_x and VOC in the presence of sunlight. Environmental stressors like land use, noise pollution, and organic and inorganic respiration are beyond the scope of this study and, hence, not considered.

The life cycle of all the pathways has four common unit processes: biomass production (UP 1), biomass transportation (UP 2), plant construction, decommissioning and disposal (UP 3), and plant operation and maintenance (UP 4). BCL and GTI pathways have hydrogen transport (UP 5) as the additional unit process. On the other hand, pyrolysis-based pathways have bio-oil transport by truck (UP 6)/pipeline (UP 7) as the additional unit process. Pyrolysis pathways involving UP 6 and UP 7 are denoted by pyrolysis I (P I) and pyrolysis II (P II), respectively, throughout the rest of this study. Fig. 2 shows the detailed system boundary for each of the pathways.

One of the differences between the system boundaries for the pyrolysis pathway compared to the BCL and GTI pathways is an additional unit operation for plant construction that is required for the fast pyrolysis of biomass feedstock. The bio-oil produced by fast pyrolysis plants is transported to a bitumen-upgrading plant for biohydrogen production. In contrast, BCL and GTI technologies produce biohydrogen directly from biomass, and later transport it for bitumen upgrading. The scope of each unit process is explained in the following sub-sections.

2.1. Scope and assumptions of unit processes

2.1.1. Biomass production

FR refers to limbs, tops, and branches left in the forest during logging operations. The current practice in Alberta is to cut trees in the stand, skid the trees to the roadside, delimb them on the roadside, and take the stem for pulp and lumber operations. Residue left in the forest are the feedstock targeted by biohydrogen plants (Kumar et al., 2003; Sarkar and Kumar, 2010a,b). This unit process involves the impact of energy and emissions related to manufacturing the forwarder and chipper, their operations, and their disposal at the end of their life. The chipper required for FR is different than that required for WF biomass, and the chipping operation is less efficient for the former (Kumar et al., 2003). As a result different chippers are considered for processing FR and WF biomass. Silviculture unit operation (replanting and nutrient replacement) is not included for producing FR-based biohydrogen. Detailed product and machinery descriptions for producing FR-based pathways and their associated unit operations are given in the Supplementary material Section (Table SM-1).

Producing whole forest biomass requires unit operations which are different from those for FR. Producing WF includes chipping, felling and skidding as unit operations. Impact due to the manufacturing and disposal of the equipment (feller, skidder, chipper) is also considered. For WF biomass, the impact from silviculture operations includes fertilizer and pesticide spraying, and fuel and machinery used for different unit operations; these are included in the base case. All the nutrients (Nitrogen – N, Phosphorous – P and Potassium – K) removed by the harvesting of WF feedstock is replaced. Impact is evaluated based on the product life cycle (i.e. energy and emissions during production of the fertilizer (N, P, K) and the pesticide including the distribution) (Kim and Dale, 2004; Borjesson, 1996). The effect of calcium replacement is ignored, however, because it is available in profuse amounts in the Western Canadian boreal forest (Kumar et al., 2003). A sensitivity case for WF biomass has been developed excluding the impact of silviculture. Details on the products and machinery involved in the utilization of WF biomass are given in the Supplementary material Section (Table SM-1).

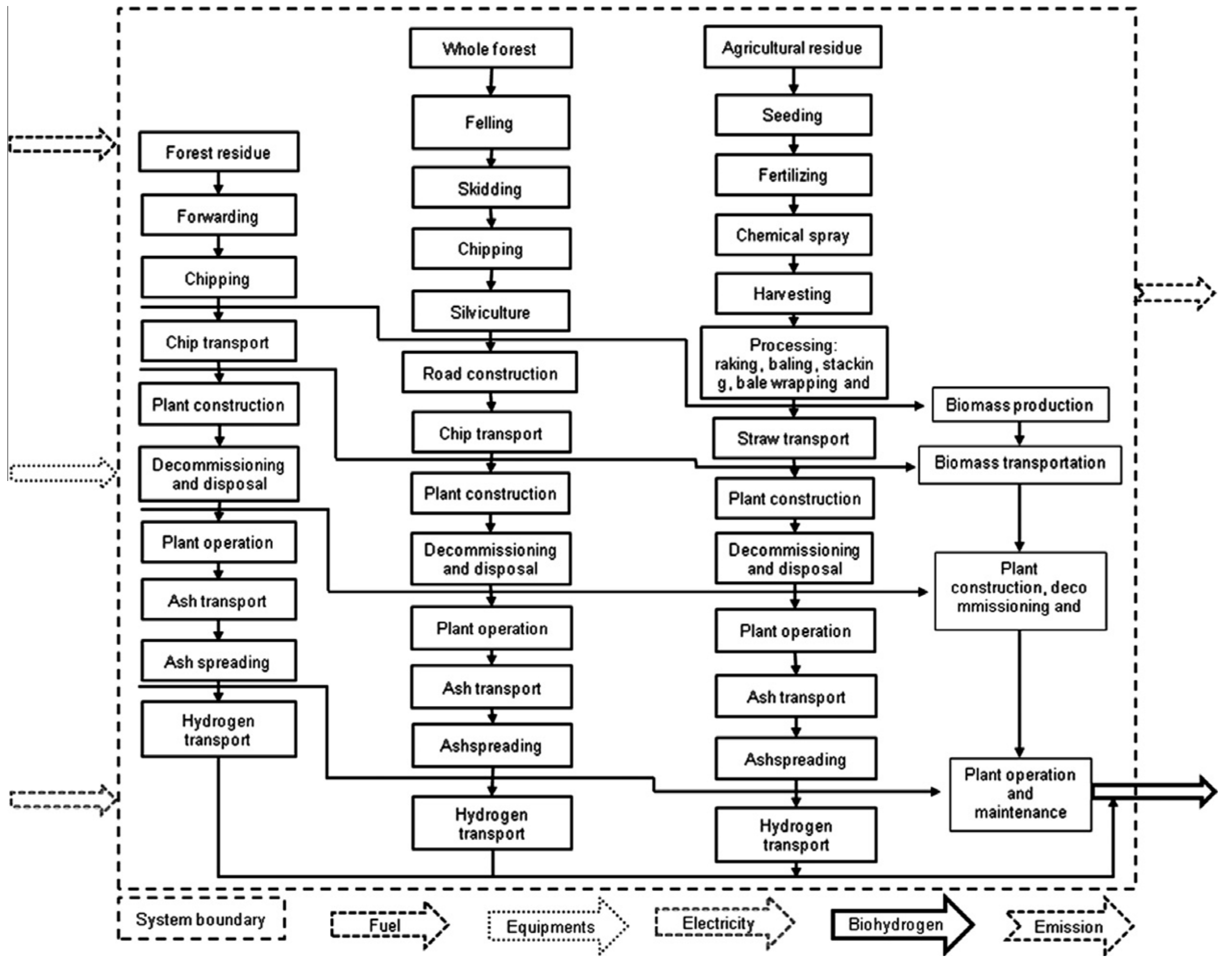


Fig. 2. System boundary for biohydrogen production pathways.

The unit operations involved in producing AR are cultivation, fertilization, chemical application, harvesting, and straw processing (including raking, baling, stacking, bale wrapping and loading). All of these unit operations are considered in the base case. The straw-to-grain ratio is assumed to be 1.1:1 on the basis of its mass fraction (Kumar et al., 2003). Accordingly, a portion of the impact from common operations for straw and grain (from cultivation to harvesting) is allocated to straw. Once again, impact is analyzed over the life cycle of fuels and equipment. Products and machinery are described in the Supplementary material Section (Table SM-1). A sensitivity case for agricultural residue has been developed that excludes the farming operations i.e. cultivation, fertilization, and chemical application. This case considers agricultural residue as waste, i.e. as having no economic value.

2.1.2. Biomass transportation

This study assumes that FR is chipped at roadside and the chips are hauled using the existing road network, hence, road construction is not included for FR chip transportation. This assumption is valid for agricultural residue bale transportation as well. In contrast, the impact from road construction is included for WF biomass in the base case. It is assumed that both WF- and FR-based chips are transported in large capacity trailer trucks. B-train chip trucks would be more appropriate for chip transport because they have a higher payload, but steep and low class logging roads act as a hindrance to the use of B-train chip trucks, making the trailer truck the only option for chip transport (MacDonald, 2006). It is assumed that similar trailer trucks are used to transport straw bales. For all feedstocks, the impact of truck manufacturing, operation and disposal is included. Products and machinery for all the unit operations related to biomass transportation are described in Section 2.2.5.

2.1.3. Plant construction, decommissioning and disposal

As stated earlier, six out of the nine pathways (BCL and GTI) require the construction of only one plant to gasify biomass and then produce biohydrogen by reforming syngas. The other three pathways require the construction of a fast pyrolysis plant in addition to a biohydrogen production plant for reforming bio-oil. The life of a plant is assumed to be 20 years in all cases. The construction material required for the different plants is estimated using data given in earlier studies (Spath and Mann, 2001; Mann and Spath, 1997). Scale factors are assumed to be 0.76, 0.68, 0.78 and 0.70, respectively, for BCL, GTI, pyrolysis and bio-oil reforming plants, and are based on detailed analyses reported in earlier studies (Sarkar and Kumar, 2010a,b). Scale factor is defined by Eq. (3) (Moore, 1959). Note that, material-embodied energy and emissions are considered over their life cycle.

$$C_i/C_o = (S_i/S_0)^n \quad (3)$$

where, C_i, C_0 = cost at size i and at reference (0) units, respectively. S_i, S_0 = size or rating of the corresponding units, and n is the scale factor. Note that the cost is a function of the amount of material used in a unit. In turn, cost is a function of a unit's volume/surface area (Moore, 1959). Thus, scale factor leads to the proposition of economies of scale (the advantage of size), and material ingestion in any equipment/unit/plant can be determined by Eq. (3) from given reference equipment/unit/plant data if the corresponding scale factor is known (Moore, 1959). For example, to build a BCL plant with a capacity of 133,125 kg H_2/day (S_0) the required amount of concrete is 10,242 tonnes (C_0) (Spath and Mann, 2001). For the same plant, the scale factor (n) is 0.76 (Sarkar and Kumar, 2010a), therefore, the concrete requirement (this study: C_i) for a BCL plant with a capacity of 250,200 kg H_2/day (this study: S_i) was estimated using Eq. (3). Likewise, the raw materials required for other pathways were estimated.

Keeping the issue of weather in mind, when roads are impassable 2 weeks of feedstock storage at the plant is considered (Kumar et al., 2003). Studies show that because stored biomass decomposes, GHG emissions may become significant if it is stored for a long period (Wihersaari, 2005). Since this study assumes the conversion of biomass is GHG-neutral and that storage is for a short time, the impact of these unit operations is ignored.

It is difficult to determine the impact from plant decommissioning because there is little reliable data, however, it is reasonable to assume that the impact from decommissioning a biomass-based plant is 3% of the plant construction (Elsayed and Mortimer, 2001). This unit process includes the cost of disposing of the construction materials. It is assumed that all the concrete and aluminum is landfilled whereas, 25% of the steel is landfilled and the rest is recycled (Spath et al., 2005; Spath and Mann, 2001). It is assumed that landfilling requires 50 km of transport using a truck similar to those used to transport biomass. Emissions related to landfilling, and to manufacturing and operating the landfilling truck are also included in this LCA. Input data for this unit process are given in Section 2.2.6.

2.1.4. Plant operation and maintenance

This unit process includes impact of feedstock preparation (drying, comminution, etc.), utilities (mainly natural gas and electricity) required in biomass conversion plants, and ash disposal. Feedstock drying is mandatory for all pathways. Before feeding into the respective reactors, the moisture content of the feedstock should be reduced to 12%, 12% and 10%, respectively, for the BCL, GTI and fast pyrolysis pathways (Spath et al., 2005; Larson et al., 2005; Bridgwater and Peacocke, 2000). Feedstock particle size is not a major concern for the BCL and GTI pathways, therefore, it is assumed that, no further size reduction is required after chipping FR and WF biomass or shredding agricultural residue. Note that, chipping is included as a unit operation in biomass production. Shredding is also included as a unit operation in plant operation and maintenance because it is done after straw is delivered to the plant in the form of bales. To ensure rapid heat transfer during pyrolysis particles should be smaller than 3 mm (Bridgwater and Peacocke, 2000), therefore, it is necessary to grind chips and straw. The impact of all these feedstock pretreatment processes is included in the analysis.

When biohydrogen is produced from FR and WF biomass using a BCL gasifier, a portion of the total plant electricity required can be supplied from electricity that the plant produces itself. Purchasing electricity from the grid is unnecessary when agricultural residue is converted into hydrogen by a BCL gasifier because the plant generates sufficient electricity for its operation (Spath et al., 2005). There is a potential for generating excess electricity from the plant which can be sold to the grid (Spath et al., 2005), but this scenario is not considered in this LCA study. Purchasing natural gas is required for all the BCL pathways even though steam is generated from waste heat and feedstock is dried using flue gases from the char combustor (Spath et al., 2005). The overall energy requirement for the BCL pathway is estimated based on a plant's energy balance. It is assumed that ash produced by the plant is disposed of 50 km away from the plant, as a complementary nutrient-replacement operation. Ash is transported by trucks used for biomass transport. Ash is spread at a rate of 1 tonne/ha using a commercial spreader (Kumar et al., 2003; Spath et al., 2005). The energy and emissions impact of disposal includes the manufacture, operation, and disposal of the truck and spreader. All are included in this LCA.

In the case of GTI pathways, the electricity produced by the plant is enough to support the feedstock pretreatment processes and other plant operations (Larson et al., 2005). Once again, credits from selling extra electricity to the grid are not considered. In addition, natural gas need not be purchased for these pathways (Larson et al., 2005). So, for GTI pathways ash disposal is the only plant operation that needs to be accounted for. Assumptions related to ash disposal are the same as these for the BCL pathways.

Neither electricity nor any fossil fuel is required to operate a biomass-based large scale fast pyrolysis plant (Ringer et al., 2006). The biomass can be dried using heat generated from the combustion of char produced by the process. Also, heat for the pyrolysis reaction can be produced by re-circulating the non-condensable exhaust gas produced during pyrolysis (Ringer et al., 2006). Assumptions regarding ash disposal are stated above. Because, bio-oil is not stable at room temperature, 10 wt.% methanol is added to it to keep its properties uniform (Sarkar and Kumar, 2010b). The impact of adding methanol is included in the analysis. Inventory data for methanol production has been taken, over the life cycle of methanol. A bio-oil and methanol blend is transported to a bitumen upgrader where the bio-oil is steam-reformed to biohydrogen. Note that, though bio-oil reforming is an endothermic process, water–gas shift reaction is an exothermic one (Spath and Mann, 2001; Sarkar and Kumar, 2010b), therefore, more steam can be produced from the exhaust of the water–gas shift reaction than was required as input for it. There is, therefore, no need for natural gas to generate steam for the water–gas shift reaction. Although this is excluded from the analysis, natural gas is required to produce steam to reform bio-oil. This steam, however, can be delivered using the existing cogeneration facility of the bitumen upgrading plant. In addition, the electricity required for running the plant equipment can be easily be produced by the cogeneration facility (Sarkar and Kumar, 2010b; Spath and Mann, 2001). Thus, in the base case it is assumed that, grid electricity and natural gas are not required to operate the reforming plant, however, diversion of thermal and electrical energy from the existing cogeneration facility of an upgrading plant to the bio-oil reforming plant may affect the overall life cycle of the pyrolysis pathways of biohydrogen production. For this reason, a sensitivity case (Table 6: case 4) has been developed which assumes that the thermal and electrical energy required for the bio-oil reforming plant are not provided by the cogeneration plant. The natural gas and electricity required for the bio-oil reforming plant are estimated to be 18.4 MJ/kg H_2 and 0.4 kWh/kg H_2, respectively (Kinoshita and Turn, 2003; Vagia and Lemonidou, 2007). Detailed inventory data for this unit process for all pathways are given in Section 2.2.7.

2.1.5. Hydrogen transport

Hydro055107gen transport is relevant to both BCL and GTI pathways. It is assumed that the hydrogen is transported for 500 km to a bitumen upgrader. Even highly compressed (50–70 MPa) hydrogen has so low a density that only 300 kg hydrogen can be carried using a conventional 36 tonne payload truck (Amos, 1998). Therefore, for distances as long as 500 km, truck transportation is an uneconomical option for hydrogen (Amos, 1998). As a result, only pipeline transport of biohydrogen is considered. The impact of manufacturing a pipeline, constructing it, and operating it, is included in this analysis. Inventory data for the unit process are given in Section 2.2.8.

2.1.6. Bio-oil transport

The distance the bio-oil will need to be transportated is assumed to be 500 km. In contrast to hydrogen transport, both pipeline and truck are considered for transportation of bio-oil and methanol blend, as both these liquids have a high density. When bio-oil and methanol blend are transported by truck, it is assumed that a high capacity B-train truck will be used. Truck transportation includes the impact from manufacturing and operating the trucks, infrastructure construction, and truck disposal. In contrast, transport of bio-oil via high-density polyethylene (HDPE) pipeline includes the impact from manufacturing the pipeline, pumps, and polyethylene foam insulators, delivering the pipeline material, constructing the pipeline, and operating it (based on an earlier study by the author, Pootakham and Kumar, 2010). Inventory data for this unit process are given in Section 2.2.9.

2.2. Inventory assessment for life cycle calculation

2.2.1. Biomass properties and plant characteristics

The yield and physical properties of biomass are very critical to performing LCA studies for biomass-based systems. These have a significant impact on various upstream and downstream operations of biomass conversion such as transportation, feedstock pretreatment, plant mass and energy balance, plant maintenance, etc. Note that, this study takes into account the higher heating value (HHV) of fuels. When assuming the yield of straw, a significant percentage (20%) of it is alloted to be left in the field to return nutrients to the soil. The biomass inventory data and general plant assumptions are given in Table 1.

2.2.2. Fuel, fertilizer, pesticide, and electricity inventory data

Fossil fuels (diesel and natural gas) are the primary energy input for almost all the unit processes. Methanol is required to stabilize bio-oil. Table 2 shows the fuels' properties, and the corresponding energy and emissions factors. Almost 82% of all the electricity generated in Alberta comes from coal-fired power plants (Environment Canada, 2009); there are, therefore, high emissions related to grid electricity. These emissions are estimated on life cycle basis and given in Table 2. The efficiency with which coal is converted to electricity is assumed to be 35%. Table 2 also shows the life cycle energy and emissions factors for different fertilizers and pesticides.

2.2.3. Material inventory data

Steel is the main raw material used in manufacturing all kinds of equipment and machines e.g. forwarders, fellers, skidders, chippers, trucks, pipelines, spreaders, etc. Concrete, along with steel and aluminum is the main raw material used to construct plants. The life cycle energy and emissions factors for different raw materials are presented in the Supplementary material Section (Table SM-2). These factors are presented for different materials over a life cycle that includes procurement, processing, transporting, usage, and disposal.

Table 1
Biomass properties and general assumptions.

Properties	FR	WF	AR	Comments/sources
Moisture content (%, wet basis)	45	50	16	These are the moisture contents of as received feedstocks. It is assumed that moisture contents wouldn't change transportation of feedstocks after preliminary processing (chipping/baling) (Sarkar and Kumar, 2009, 2010a).
Bulk density (kg/m^3)	230	250	175	These values correspond to wet bulk density of as received feedstocks (Angus-Hankin et al., 1995; Sarkar and Kumar, 2010a).
Higher heating value (MJ/dry kg)	19.7	20	16.3	Sarkar and Kumar (2009, 2010a)
Ash content (%)	3	1	4	Kumar et al. (2003)
Biomass yield (dt/hectare)	0.247	84	0.333	Yield of WF is based on mixed stands of hardwood and spruce in Alberta for 100-year rotation of forest growth (Kumar et al., 2003). Yield of FR is based on the assumption that 20% of the whole tree consists of forest residues (Kumar et al., 2003).
Plant operating factor				These are conventional operating factors being used for biomass based plants (Kumar et al., 2003; Sarkar and Kumar, 2009, 2010a,b).
Year 1	0.7	0.7	0.7	
Year 2	0.8	0.8	0.8	
Year 3 onwards	0.85	0.85	0.85	
Average hauling distance				Hauling distance is related to the plant size and net biomass yield. Geometric and tortuosity factor was assumed 1 and 1.27, respectively while modeling biomass harvesting area requirement (Overend, 1982). Harvesting area for agricultural residue is assumed as a square and agricultural residue fed gasification/pyrolysis plant will be located at the intersecting point of the diagonals. In contrast, harvesting area for FR and WF biomass is assumed circular and respective biohydrogen/pyrolysis plants are assumed to be located at the center of the circle. This methodology is already used in earlier studies (Sarkar and Kumar, 2009, 2010a). Note that, bitumen upgrading plant is located 500 km away from gasification/pyrolysis plant. Biohydrogen from gasification plant is transported by pipeline to an upgrader and bio-oil from pyrolysis plant is transported by pipeline or truck to the upgrading plant to produce biohydrogen.
BCL pathways				
Year 1 (km)	84	22.4	102	
Year 2 (km)	90	22.4	109	
Year 3 onwards (km)	93	22.4	113	
GTI pathways				
Year 1 (km)	84	25.8	102	
Year 2 (km)	90	25.8	109	
Year 3 onwards (km)	93	25.8	113	
Pyrolysis pathways				
Year 1 (km)	69	18.3	83	
Year 2 (km)	73	18.3	89	
Year 3 onwards (km)	76	18.3	92	
Biohydrogen yield				Biohydrogen yield from BCL and GTI pathways are taken from Sarkar and Kumar (2009, 2010a). Bio-oil yield[a] from pyrolysis pathways is assumed 70.7 wt.% (10.8 wt.% water), 50.53 wt.% (20.57 wt.% water), and 42 wt.% (19.07 wt.%) from WF, FR and AR biomass, respectively. Biohydrogen yield from bio-oil reforming is assumed to be 13.92 wt.% of water free bio-oil (Sarkar and Kumar, 2010).
BCL (kg H_2/dt)	83.4	83.4	83.4	
GTI (kg H_2/dt)	83.4	83.4	83.4	
Pyrolysis[a] (kg H_2/dt)	41.7	83.4	31.9	

[a] Higher ash and fixed carbon content results in lower bio-oil yield during fast pyrolysis (Bridgwater and Peacocke, 2000; Sarkar and Kumar, 2010b). Ash content of WF, FR, and AR feedstock is 1%, 3%, and 4% respectively (Kumar et al., 2003). In contrast, fixed carbon content is found to be 17%, 19.59%, 18–21% respectively (Vamvuka et al., 2003; McKendry, 2002). Unlike ash and fixed carbon, volatile matters in biomass, favors the bio-oil production. Volatile matters in WF, FR, and AR are found to be 82%, 79.8%, and 46–59% respectively (Vamvuka et al., 2003; McKendry, 2002). Cellulose and hemi-cellulose are the other important parameters in determining the bio-oil yield. Higher content of them in WF and FR (35–50% cellulose, 20–30% hemi-cellulose) compared to AR (33–40% cellulose, 20–25%) result in better bio-oil yield (McKendry, 2002).

Table 2
Energy input/output ratio and emission factors for electricity, different fuels and chemicals.

Category	Diesel [a]	Natural gas [a]	Methanol [a]	Electricity [b] (unit/MWh)	Fertilizer [c] (Unit/kg)			Pesticide [c] (unit/kg)	Comments/sources
					N	P	K		
HHV (MJ/kg)	46.03	49.1	22.7	–	–	–	–	–	Fuel, energy and emission factors are determined based on the published literature (($(S\&T)^2$ Consultants Inc. 2010; Spath and Mann, 2000; Furuholt, 1995; Bernesson et al., 2004). Fuel heating values and densities are taken from Environment Canada (2009). It is assumed that methanol used to stabilize bio-oil is derived from biomass. Energy and emission factors of Alberta are estimated from earlier studies (Cuddihy et al., 2005; Environment Canada, 2009). Fertilizer and pesticide factors are derived from literature (Kim and Dale, 2004; Borjesson 1996).
Density (kg/m^3)	832	0.78	792	–	–	–	–	–	
kg CO_{2eq}/GJ	94.2	56.6	16	820	3.27	1.34	0.64	24.5	
kg SO_{2eq}/GJ	0.37	0.13	2E−03	0.57	0.38	0.40	0.40	2.96	
kg (NO_x + VOC)/GJ	0.59	0.22	1E−03	0.585	0.40	0.41	0.41	3.01	
GJ/GJ	1.22	1.11	0.04	2.86	0.05	0.01	0.004	0.12	

[a] All of the factors are estimated based on the respective life cycle of the fuel. Impact factors for utilization of diesel include impacts from operations like dispensing, distribution and storage, production, transmission, land use change, feedstock recovery, leaks, flares and combustion. Natural gas and methanol impacts factors also include these operations. All the values are given as per GJ of fuel content.
[b] Energy and emission impacts are corresponding to 1 MWh electricity generation and estimated based on the life cycle of Alberta grid.
[c] Energy and emission impacts are based on the life cycle of 1 kg fertilizer/pesticide production.

2.2.4. Inventory data for biomass production

The machinery and products needed for biomass (FR, WF, and AR) production are described in the Supplementary material Section (Table SM-1).

2.2.5. Inventory data for biomass transportation

It is assumed that biomass will be transported using the high capacity trailer trucks. A trailer truck is capable of carrying 23 wet tonnes of biomass per trip. Each truck manufactured out of 14 tonnes of steel. A typical fuel efficiency of 6 miles per gallon and 5 miles per gallon is assumed for trailers is traveling without and with load, respectively (Mann and Spath, 1997).

Note that it is difficult to determine the exact amount of road construction required for WF biomass hauling. In forests, two types of roads are required, namely, primary and secondary roads. Secondary roads are required for the operation of forwarders, fellers and skidders. Whole trees are skidded to the primary roadside and chipped; the chips are then transported by truck to the plant. Quality is not a major concern with secondary roads because equipment like fellers/skidders operate at low speed. In contrast, primary roads are as good as highways. The impact from secondary road construction is not considered in this study for two reasons. First, there is a scarcity of reliable data, and, second, overall length is negligible compared with primary roads. Based on the optimal road spacing design for forests provided to us in discussion with an expert (Smyl Fulton, Weyerhaeuser Corporation, 2011), it is estimated that over 20 years, 1050, 1395, and 700 km of primary road network (6 m wide) are required, respectively for the BCL, GTI and pyrolysis pathways of producing biohydrogen from WF feedstock (Demir and Ozturk, 2004). Energy and emissions factors for road construction are 1731 GJ/km, 403,845 kg CO_{2eq}/km, 1015 kg SO_{2eq}/km, and 1155 kg (NO_x + VOC)/km, respectively (Stripple, 2001). A sensitivity case has been developed which excludes road construction for WF biomass.

2.2.6. Inventory data for plant construction, decommissioning, and disposal

The steel, concrete and aluminum required to construct a BCL plant construction (for all the feedstocks) are 5350, 16,535, and 44 tonnes, respectively. To construct a GTI plant (for processing FR and AR), the material required is 5084, 15,720, and 42 tonnes, respectively. A WF-based GTI plant requires 6182, 19,120, and 50 tonnes of materials, respectively (Spath and Mann, 2001; Mann and Spath, 1997).

A FR-based pyrolysis and bio-oil reforming plant together requires 6285, 19,435, and 52 tonnes of steel, concrete and aluminum, respectively. WF-based plants require 7775, 24,040, and 64 tonnes of materials, respectively. Agricultural-residue-based plants require 5880, 18,175, and 48 tonnes of materials, respectively (Spath and Mann, 2001; Mann and Spath, 1997).

For all plants, the decommissioning impact is assumed to amount to 3% of the construction impact (Elsayed and Mortimer, 2001). After decommissioning, materials are transported by truck for 50 km to a landfill. Inventory data for the truck are given above. Emissions from landfilling steel is 0.01 tCO_{2eq}/t of material; for concrete it is 0.044 tCO_{2eq}/t of material (ICF Consulting, 2005; EPA, 2010). There is too little data available on other factors such as energy, ARP, and GOP inventory so these are not considered for landfilling operations. They are assumed to be negligible.

2.2.7. Inventory data for plant operation and maintenance

The natural gas required to produce biohydrogen using a BCL gasifier has been found to be 0.38 m^3/kg H_2 for all three feedstocks (Spath et al., 2005). The grid electricity that must be purchased for forest residue and WF-based BCL gasifiers is 1.14 kWh/kg H_2 and 1.48 kWh/kg H_2 produced, respectively (Spath et al., 2005), however, agricultural-residue-based biohydrogen production using a BCL gasifier does not require any electricity be purchased (Spath et al., 2005). Neither natural gas nor electricity purchases are required for GTI-gasifier-based biohydrogen production (Larson et al., 2005). No fossil fuel or electricity need be purchased to operate a fast pyrolysis plant (Ringer et al., 2006). Methanol (10 wt.%) is needed for bio-oil stabilization. Inventory data for methanol have been given above. Bio-oil reforming plants also do not require the purchase of natural gas or electricity purchase (Sarkar and Kumar, 2010b). Ash is disposed 50 km away from the plant and is spread (1 tonne ash/ha) to replace nutrients. The ash content in bio-oil is less than 0.1% (Bridgwater and Peacocke, 2000), hence, the impact from ash disposal is ignored for bio-oil reforming plants. The inventory data for trucks used to transport in ash is as stated above. It is assumed that 40′ fertilizer spreaders will be used to spread ash. The productivity, fuel consumption rate, and lifetime of a spreader is estimated to be 4.4 ha/hr, 5 L/hr, and 1200 h respectively (Mann and Spath, 1997).

2.2.8. Inventory data for biohydrogen transport

At normal temperature and pressure, the density of hydrogen is very low (0.089 kg/m^3) (Spath and Mann, 2001), therefore, hydrogen must be highly compressed to make transportation cost-effective. As noted earlier, compressing hydrogen to a pressure as high as 70 MPa, does not improve its density significantly. Pipelines are the most viable option for transporting hydrogen over a distance of 500 km. Hydrogen from the BCL and GTI pathways is available in compressed form at 7 and 6 MPa, respectively (Spath et al., 2005; Larson et al., 2005). Biohydrogen produced by a GTI gasifier can easily be compressed to 7 MPa. It is not necessary to purchase grid electricity because plants produce sufficient amounts of surplus electricity. After hydrogen is separated from syngas, residual syngas (typically 5%) from the pressure swing adsorption (PSA) unit is burned in a boiler to operate a Rankine cycle; in this way extra electricity is generated (Larson et al., 2005). Hydrogen can be directly fed into a pipeline at 7 MPa (Spath et al., 2005). To ensure the comparability of the pathways, pipeline systems are designed so that hydrogen will be delivered to the upgrading plant at 2.5 MPa. Inventory data are given below for transporting biohydrogen by pipeline.

2.2.9. Pipeline characteristics

For BCL pathways 12 in. is considered, the nominal pipe size; the pipeline is made of ASTM A 106 (grade B steel). The same material and pipe size are used for forest- and agricultural-residue-based GTI gasification, however, WF-fed GTI plants, though the material is the same, require a 14 in. pipe. Pipeline material is selected from Mohitpour et al. (2007). Nominal pipe size is selected from GPSA (1972).

2.2.10. Pipeline manufacturing and construction

The steel required for 12 and 14 in. pipelines are 97.5 and 107.3 kg/m of pipeline, respectively (GPSA, 1972). Pipeline weight is estimated for a wall thickness of 0.5 in. The diesel consumed during pipeline construction is 3612.5 L/km of pipeline (Pootakham and Kumar, 2010).

2.2.11. Pipeline operation

The pressure drops for 12 and 14 in. pipelines are 18 and 31.8 kPa/km, respectively. As a result, one booster station is required for the former and three booster stations are required for the latter in order to supply hydrogen at 2.5 MPa to the upgrader. The former requires 186 MWhr per day of electrical energy, and the latter requires 491 MWhr per day of electrical energy for pipeline operations. Pressure drops were estimated from Mohitpour et al. (2007).

2.2.12. Bio-oil transport inventory data

Unlike biohydrogen, bio-oil has a high density of 1200 kg/m^3. Hence trucks and pipelines are considered as two modes of transportation. It is assumed that bio-oil and methanol blend will be transported either using B-train trucks of 60 m^3 capacity or an HDPE pipeline bitumen if the distance to bitumen upgrading plant is 500 km or more. Inventory data for bio-oil transportation are presented in Table 3.

3. Result and discussion

3.1. Life cycle energy impact of BCL pathways

The energy impact and NER corresponding to the functional unit for different BCL pathways are shown in Table 4. Note that, in order to determine NER, the HHV of hydrogen has been assumed to be 142 MJ/kg (Spath and Mann, 2001). It is evident that for the FR-and WF-based BCL pathways, plant operation and maintenance (UP 4) contributes significantly to the overall energy impact. This unit process is responsible for 65% and 63% of the total energy impact for the two biomass feedstocks, respectively. The key reason is the high requirement for electricity and natural gas to run the plant. On the other hand, for AR, biomass production (UP 1) (54%) has more impact than plant operation and maintenance (UP 4) (29%). There are two reasons for this. First, electricity need not be purchased to run the plant. Second, too many upstream unit operations are involved during AR production. Transporting biomass (UP 2) and biohydrogen (UP 5) together affect the overall energy impact by 6%, 4%, and 5.5% for AR, FR, and WF feedstock, respectively. The impacts from plant construction, decommissioning, and disposal (UP 3) are found to be negligible compared to other unit processes for all the feedstocks. Among the feedstocks, FR to biohydrogen has the highest NER – 3.3. The details of the life cycle energy impact for different BCL pathways are given in Table 4.

3.2. Life cycle energy impact of GTI pathways

Unlike for BCL pathways, for FR- and WF-based GTI pathways, hydrogen transportation (UP 5) is the main contributor to the overall energy impact. This unit process contributes 45% and 58% of the total energy impact, respectively, for the two feedstocks. Compared to FR-, WF-based GTI plants have a higher impact from transporting hydrogen. WF-based GTI plants have an optimum capacity of 4000 dtpd, compared to 3000 dtpd for FR. The designed pipeline size was found to be 12 and 14 in., respectively, for FR and WF pathways, and the pressure drops were 18 and 31.8 kPa/km for the respective pipeline sizes. As a result, three booster stations

Table 3
Inventory data for bio-oil transport modes.

Mode	Category	Values	Comments/Sources
Truck	Energy impact Emission impact	0.85 MJ/m^3/km 56 gmCO$_{2eq}$/m^3/km 0.23 gmSO$_{2eq}$/m^3/km 0.36 gm(NO$_x$ + VOC)/m^3/km	Impacts include truck manufacturing, infrastructure construction, and truck operation. Energy and GHG emission impact are adjusted for biohydrogen plant (Pootakham and Kumar, 2010). The authors evaluated other impacts based on the material inventory data from Pootakham and Kumar (2010).
Pipeline	Energy impact Emission impact	3.77 MJ/m^3/km 0.31 kgCO$_{2eq}$/m^3/km 0.21 kgSO$_{2eq}$/m^3/km 0.21 kg(NO$_x$ + VOC)/m^3/km	Impacts include HDPE pipeline manufacturing, pump manufacturing, polyethylene insulator manufacturing, truck delivery of pipeline construction materials, pipeline construction, and pipeline operation. Energy and GHG emission impact are adjusted for biohydrogen plant (Pootakham and Kumar, 2010). The authors evaluated other impacts based on the material inventory data from Pootakham and Kumar (2010)

Table 4
Life cycle energy performance of biohydrogen pathways.

Feedstock	UP	MJ/kg H_2			
		BCL	GTI	P I	P II
AR	UP 1	30.6	30.6	79.8	79.8
	UP 2	2.9	2.9	6.2	6.2
	UP 3	0.13	0.12	0.6	0.6
	UP 4	16.2	0.1	1.3	1.3
	UP 5	6.9	6.9	–	–
	UP 6	–	–	5	–
	UP 7	–	–	–	23.5
	Overall energy impact, MJ/kg H_2 (NER)	57 (2.5)	40.6 (3.5)	92.8 (1.6)	111 (1.3)
FR	UP 1	4.6	4.6	9.3	9.3
	UP 2	3.7	3.7	6	6
	UP 3	0.13	0.12	0.9	0.9
	UP 4	27.8	0.05	1.1	1.1
	UP 5	6.9	6.9	–	–
	UP 6	–	–	4.6	–
	UP 7	–	–	–	21.6
	Overall energy impact, MJ/kg H_2 (NER)	43.1 (3.3)	15.4 (9.3)	21.9 (6.5)	38.9 (3.7)
WF	UP 1	9.4	9.4	9.4	9.4
	UP 2	2.2	2.3	2	2
	UP 3	0.13	0.12	0.3	0.3
	UP 4	31.3	0.02	0.7	0.7
	UP 5	6.8	16.3	–	–
	UP 6	–	–	3.2	–
	UP 7	–	–	–	15.1
	Overall energy impact, MJ/kg H_2 (NER)	49.8 (2.9)	28.1 (5)	15.6 (9.1)	27.5 (5.2)

BCL = Battelle Columbus Laboratory, GTI = Gas Technology Institute, P I: pyrolysis I pathway (includes UP 1–UP 4 and UP 6), and P II: pyrolysis II pathway (includes UP 1–UP 4 and UP 7). UP 1: unit process 1 (biomass production), UP 2: unit process 2 (biomass transportation), UP 3: unit process 3 (plant construction, decommissioning, and disposal), UP 4: unit process 4 (plant operation and maintenance), UP 5: unit process 5 (hydrogen transport by pipeline), UP 6: unit process 6 (bio-oil transport by truck), and UP 7: unit process 7 (bio-oil transport by pipeline). AR = agricultural residue, FR = forest residue, and WF = whole forest.

are required for WF feedstock compared to one for FR if biohydrogen is to be delivered to the bitumen upgrader at 2.5 MPa. As a great deal of electricity is required during hydrogen compression, the overall impact is significant for WF feedstock. In addition, the material (steel) required for 12 and 14 in. pipelines was found to be 97.5 and 107.3 kg/m, respectively, for FR and WR cases; this also had an impact in the two cases. For AR-based GTI plants, biomass production (UP 1) (75%) supplies the largest share of the overall energy impact. For this feedstock impact from transporting hydrogen (UP 5) is found to be 17% of the total. In all cases, NER is higher for GTI pathways than for BCL pathways. This is because, for the GTI-based plant, neither grid electricity nor natural gas need be purchased to run the plant; therefore, the impacts from GTI plant operation and maintenance (UP 4) are also negligible. As with BCL plants, the impact from GTI plant construction, decommissioning, and disposal (UP 3) has been found to be insignificant regardless of the feedstock. NERs for GTI plants are higher for all the feedstocks compared to their respective BCL plants. FR-based GTI plants exhibit the highest NER – 9.3. The details of the life cycle energy impacts for different GTI pathways are given in Table 4.

3.3. Life cycle energy impact for pyrolysis pathways

Table 4 shows the life cycle energy impact of different pyrolysis pathways for producing biohydrogen. NERs for the AR pathway are significantly lower than for the FR and WF pathways. The key reason for this is the lower bio-oil yield (42 wt.%) from AR feedstock compared to FR (50.53 wt.%) and WF (70.7 wt.%) feedstocks. The lower bio-oil yield from AR translates into more upstream operations during biomass production (UP 1) and transportation (UP 2). Note that, the optimum capacity for pyrolysis pathways was 2000 dtpd for all the feedstocks. In contrast to AR, the higher bio-oil yields from WF and FR biomass result in improved energy efficiency for the respective pathways. Bio-oil transportation by pipeline (UP 7) is too expensive in terms of energy consumption (by truck it is UP 6) because compressing bio-oil is very energy intensive. As with the BCL and GTI pathways, plant construction, decommissioning and disposal (UP 3) have a minor impact on the life cycle energy levels of pyrolysis pathways. The WF-feedstock-based pathway has the highest NER for both P I (bio-oil transported by truck) and P II (bio-oil transported by pipeline); these are 9.1 and 5.2, respectively. In brief, for biohydrogen production pathways, NERs lie in the range of 1.3–9.3. In contrast, coal- and natural-gas-based hydrogen production plants demonstrate NERs in the range of 0.57–0.67 (Spath and Mann, 2001; Ruether et al., 2005).

3.4. Life cycle emission impact

The life cycle GHG emissions from the BCL pathways are shown in Fig. 3. As the figure shows, plant operation and maintenance (UP 4) is the main contributor to the overall life cycle GHG emissions from the FR- and WF-based BCL pathways; it accounts for 60% and 54% of the total life cycle GHG emissions from the respective pathways. The key reason for this is the high requirement for electricity and natural gas to operate the respective plants. In contrast, the AR-based BCL pathway, biomass production (UP 1) (57%) has a higher impact on the life cycle GHG emissions than does plant operation and maintenance (UP 4) (22%). There are two major reasons for this. First, no electricity purchase is required to run the plant. Second, there are too many upstream unit operations involved in producing the AR biomass. Transportation of biomass (UP 2) and biohydrogen (UP 5) together contributes 20%, 28%, and 24% to the overall GHG emissions impact for the AR-, FR-, and WF-based BCL pathways, respectively. As transportation of biohydrogen (UP 5) involves high electricity consumption in booster station(s), it is more GHG-emissions intensive than biomass transportation (UP 2). Irrespective of the BCL pathways considered, the impact of GHG emissions from plant construction, decommissioning, and disposal (UP 3) are negligible.

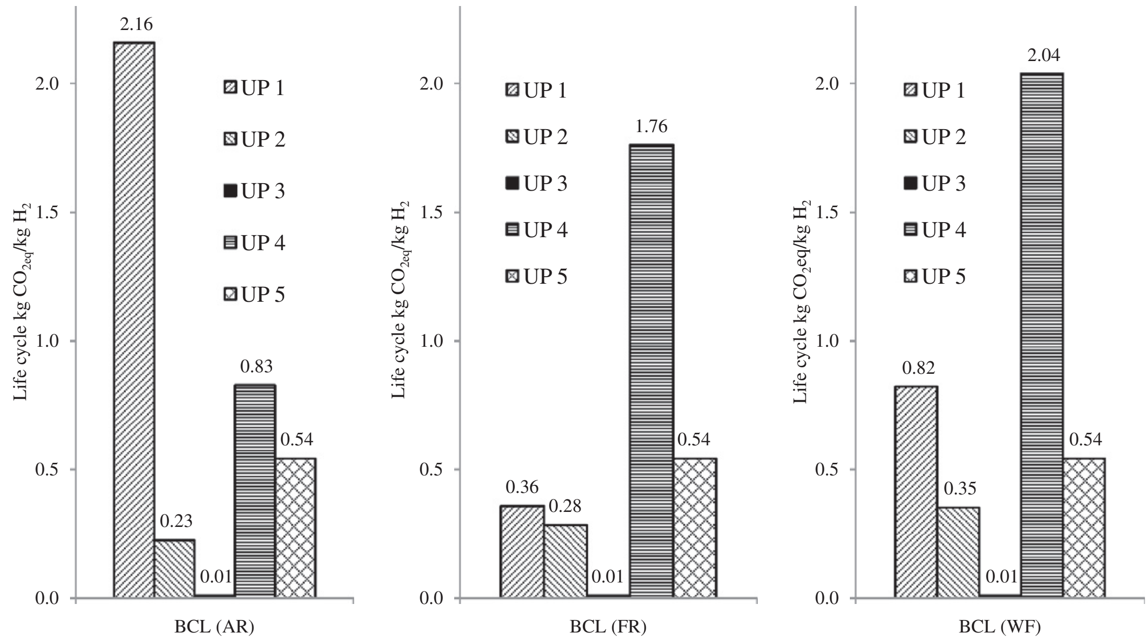


Fig. 3. Life cycle GHG emissions impacts from Battelle Columbus Laboratory (BCL) pathways. UP 1: unit process 1 (biomass production), UP 2: unit process 2 (biomass transportation), UP 3: unit process 3 (plant construction, decommissioning, and disposal), UP 4: unit process 4 (plant operation and maintenance), and UP 5: unit process 5 (hydrogen transport by pipeline). AR = agricultural residue, FR = forest residue, and WF = whole forest.

Life cycle GHG emissions from the GTI pathways are shown in Fig. 4. As found in Fig. 4, biomass production (UP 1) and biohydrogen transportation by pipeline (UP 5) are the main contributors to the overall GHG emissions from the AR-based GTI pathway. For this pathway, the former and the latter unit processes are responsible for 73% and 18% of the overall GHG emissions, respectively. Biomass preprocessing operations and high electricity consumption are the main sources of GHG emissions. For the FR-based GTI pathway, biomass production (UP 1), transportation (UP 2), and pipeline transport of biohydrogen (UP 5) contribute 30%, 23%, and 45% of the overall GHG emissions, respectively. In contrast, for the WF-based GTI pathway these unit processes account for 33%, 15%, and 52% of the life cycle GHG emissions, respectively. The impact from plant construction, decommissioning, and disposal (UP 3), and plant operation and maintenance (UP 4) are found to be negligible for all the GTI pathways.

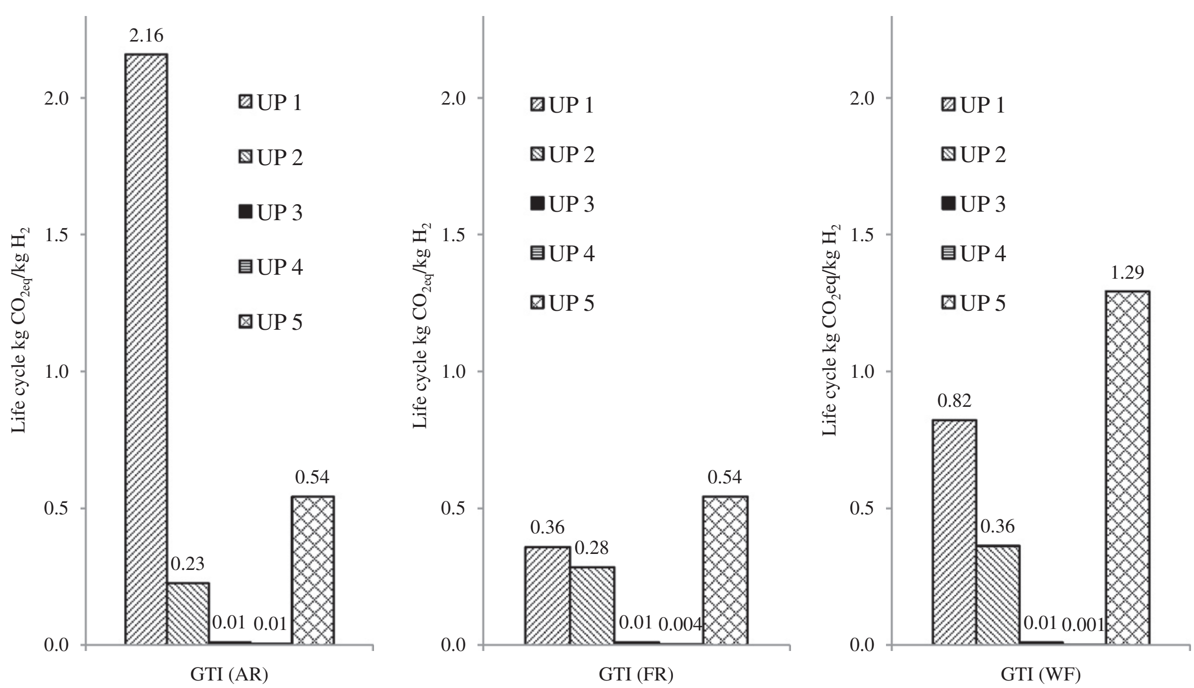


Fig. 4. Life cycle GHG emissions impacts from Gas Technology Institute (GTI) pathways. UP 1: unit process 1 (biomass production), UP 2: unit process 2 (biomass transportation), UP 3: unit process 3 (plant construction, decommissioning, and disposal), UP 4: unit process 4 (plant operation and maintenance), and UP 5: unit process 5 (hydrogen transport by pipeline). AR = agricultural residue, FR = forest residue, and WF = whole forest.

Fig. 5 describes the life cycle GHG emissions impact from the pyrolysis pathways. For the AR-based P I pathway, biomass production (UP 1) accounts for 86% of the overall GHG emissions. Once again, preprocessing operations in UP 1 play the key role. In contrast, for the AR-based P II pathway, biomass production (UP 1) and pipeline transport of bio-oil (UP 7) contribute 69% and 24% of the overall GHG emissions, respectively. Pipeline transport of bio-oil (UP 7) is the most expensive unit process in terms of GHG emissions for the FR- (58%) and WF-based (51%) P II pathways. On the other hand, biomass production (UP 1) contributes 46% and 58% of the overall GHG emissions from the FR- and WF-based P I pathways, respectively. Note that, the impact from plant construction, decommissioning, and disposal (UP 3), and from plant operation and maintenance (UP 4) is negligible for all the pyrolysis (P I/P II) pathways irrespective of the feedstock.

Fig. 6 compares the life cycle GHG emissions of all the biohydrogen production pathways considered in this study. The FR-based GTI pathway emits the least amount of GHGs (1.20 kg CO_{2eq}/kg H_2) among the pathways. In contrast, the AR-based pyrolysis (II) pathway emits the greatest amount (8.12 kg CO_{2eq}/kg H_2). Among

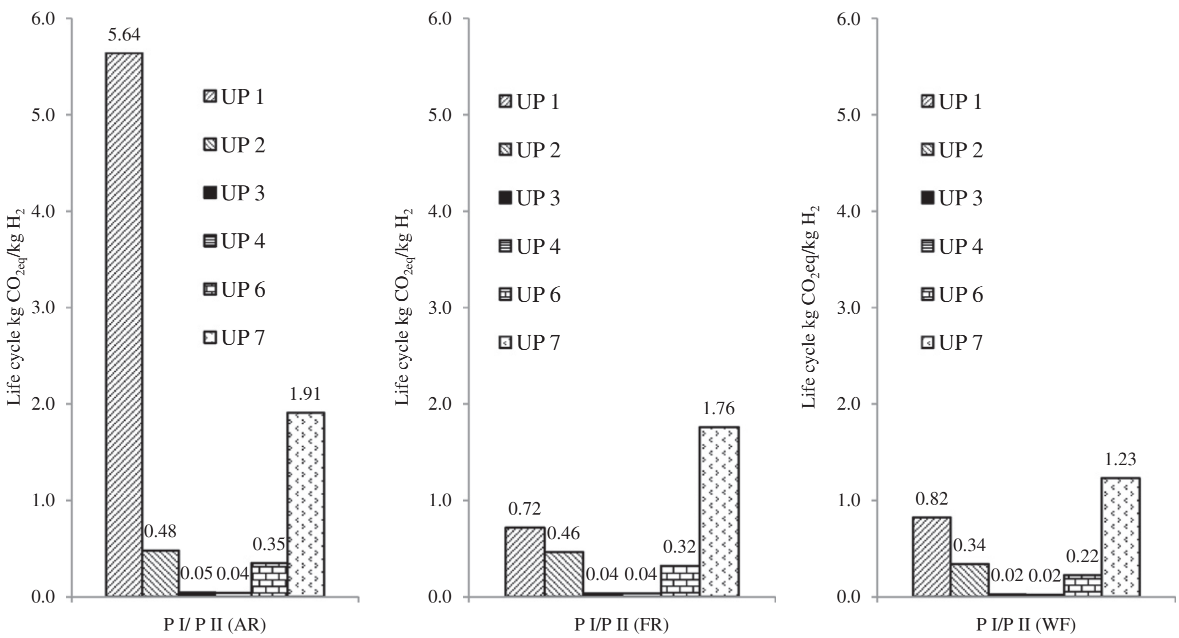


Fig. 5. Life cycle GHG emissions impacts from pyrolysis pathways. P I: pyrolysis I pathway (includes UP 1–UP 4 and UP 6) and P II: pyrolysis II pathway (includes UP 1–UP 4 and UP 7). UP 1: unit process 1 (biomass production), UP 2: unit process 2 (biomass transportation), UP 3: unit process 3 (plant construction, decommissioning, and disposal), UP 4: unit process 4 (plant operation and maintenance), UP 6: unit process 6 (bio-oil transport by truck), UP 7: unit process 7 (bio-oil transport by pipeline). AR = agricultural residue, FR = forest residue, and WF = whole forest.

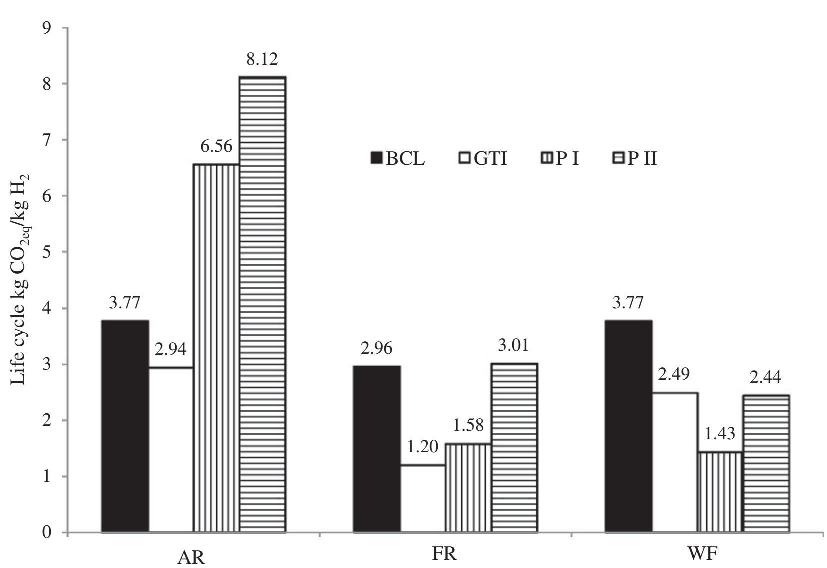


Fig. 6. Comparison of life cycle GHG emissions intensities of biohydrogen production pathways. BCL: Battelle Columbus Laboratory (includes UP 1–UP 5), GTI: Gas Technology Institute (includes UP 1–UP 5), P I: pyrolysis I pathway (includes UP 1–UP 4 and UP 6), and P II: pyrolysis II pathway (includes UP 1–UP 4 and UP 7). UP 1: unit process 1 (biomass production), UP 2: unit process 2 (biomass transportation), UP 3: unit process 3 (plant construction, decommissioning, and disposal), UP 4: unit process 4 (plant operation and maintenance), UP 5: unit process 5 (hydrogen transport by pipeline). UP 6: unit process 6 (bio-oil transport by truck), UP 7: unit process 7 (bio-oil transport by pipeline). AR = agricultural residue, FR = forest residue, and WF = whole forest.

the BCL pathways, FR-based hydrogen emits the fewest GHGs (2.96 kg CO_{2eq}/kg H_2). Life cycle GHG emissions from AR- and WF-based biomass are 27.4% and 27% higher than that, respectively. The life cycle GHG emissions from the GTI pathways of AR and WF are 145% and 108% higher than those from the FR pathway (1.20 kg CO_{2eq}/kg H_2). Because of the low bio-oil yield from AR, GHG emissions intensities from its P I (6.56 kg CO_{2eq}/kg H_2) and P II (8.12 kg CO_{2eq}/kg H_2) pathways is significantly higher than that from the other feedstocks.

Life cycle ARP and GOP impact is broken down in Table 5. Among the BCL pathways, the lowest ARP (6.23E−03 kg SO_{2eq}/kg H_2) is found for FR-based hydrogen. The impact from WF- and AR-based hydrogen are 26% and 75% higher than that, respectively. As with ARP, the lowest GOP (8.56E−03 kg [NO_x + VOC]/kg H_2) among the BCL pathways belongs to FR. The impacts from WF- and AR-based hydrogen are 25% and 93% higher than that, respectively. As with BCL, FR is found to be the best alternative for biohydrogen production among the GTI pathways from the perspective of ARP and GOP. On the other hand, among the pyrolysis pathways (P I/P II) the lowest ARP and GOP levels are found for WF-based hydrogen generation. For P I, these are 5.11E−03 kg SO_{2eq}/kg H_2 and 7.42E−03 kg (NO_x + VOC)/kg H_2, respectively. In contrast, for P II, these are 5.16E−03 kg SO_{2eq}/kg H_2 and 7.02E−03 kg (NO_x + VOC)/kg H_2. The results are concisely presented in Table 6 under the base case. Note that, variations in the levels of ARP and GOP emissions with different feedstocks and pathways strongly follow variations in their respective energy and GHG emissions. They can thus be explained based on the same argument presented earlier.

Based on this LCA study, GHG, ARP, and GOP emissions from producing biohydrogen fall in the range of 1.2–8.1 kg CO_{2eq}/kg H_2, 3.72E−03 to 2.25E−02 kg SO_{2eq}/kg H_2, and 4.69E−03 to 3.49E−02 kg (NO_x + VOC)/kg H_2. In contrast, emissions from producing coal- and natural gas-based hydrogen fall in the range of 12–43 kg CO_{2eq}/kg H_2, 2.0E−02 to 1.0E−01 kg SO_{2eq}/kg H_2, and 3.0E−02 to 2.0E−01 kg (NO_x + VOC)/kg H_2 (Koroneos et al., 2005; Spath and Mann, 2001; Ruether et al., 2005; Dufoura et al., 2009). Note that, for all the biohydrogen production pathways emissions levels are significantly less than those from fossil-fuel-based hydrogen (Figs. 3–5, Tables 5 and 6). Emissions from AR-based hydrogen are comparatively higher than those from FR- and WF-based hydrogen. UP 1 is predominantly responsible for that, because the UP 1 of AR-based hydrogen involves more operations in biomass farming, harvesting, and processing (Supplementary material Section Table SM-1). Due to the lower bio-oil yield from AR compared to FR and WF feedstock, the influence of UP 1 is even more significant in the case of the P I and P II pathways (Fig. 5, and Tables 5 and 6).

3.5. Sensitivity analysis

A sensitivity analysis with various possible scenarios has been performed in this study. The parameter was changed in each scenario independently of the others so the results could be compared with those from the base case. Results from the sensitivity analysis are summarized in Table 6. Case 1 considers AR as waste; therefore, farming operations like seeding, fertilizing, chemical spraying, etc. are excluded from the system boundary of AR-based conversion pathways, though processing operations like harvesting, raking, baling, wrapping, stacking, etc. are included. As suggested by Case 1, farming operations have a huge impact on the overall energetic and environmental performance of AR-based hydrogen. The overall impact of all AR-based pathways drops significantly due to the exclusion of the farming operations. Grid emissions for Alberta are likely to fall if coal is replaced with renewables and other relatively environment-friendly alternatives for power generation. Hence, Case 2 develops a 'what if scenario' involving a 20% drop in grid emissions intensity. As shown in Case 2, a 20% reduction in grid emissions changes the results noticeably, though less significantly than the former case. Case 3 suggests that, excluding silviculture and road construction from WF-based biomass reduces the impact significantly compared to all the pathways. Case 4 has been developed assuming that, energy required in the bio-oil reforming plant will be purchased instead of provided by waste

Table 5
ARP and GOP impacts from biohydrogen pathways.

Feed	Unit process	kg SO_{2eq}/kg H_2			kg (NO_x + VOC)/kg H_2		
		BCL	GTI	P I/P II	BCL	GTI	P I/P II
FR	UP 1	1.4E−03	1.4E−03	2.9E−03	2.2E−03	2.2E−03	4.4E−03
	UP 2	1.2E−03	1.2E−03	1.9E−03	1.7E−03	1.7E−03	2.9E−03
	UP 3	8.5E−05	8.0E−05	3.0E−04	4.4E−05	4.2E−05	1.5E−04
	UP 4	2.5E−03	1.6E−05	8.7E−05	3.9E−03	2.4E−05	7.5E−05
	UP 5	1E−03	1E−03	–	6.6E−04	6.6E−04	–
	UP 6	–	–	1.3E−03	–	–	2.1E−03
	UP 7	–	–	1.4E−03	–	–	1.5E−03
WF	UP 1	3.0E−03	3.0E−03	3.0E−03	4.7E−03	4.7E−03	4.7E−03
	UP 2	1.0E−03	1.1E−03	9.5E−04	1.3E−03	1.3E−03	1.2E−03
	UP 3	8.5E−05	7.3E−05	1.8E−04	4.4E−05	3.8E−05	9.6E−05
	UP 4	2.7E−03	5.3E−06	4.4E−05	4.1E−03	7.9E−06	2.7E−05
	UP 5	1E−03	1.5E−03	–	6.7E−04	1.2E−03	–
	UP 6	–	–	9.1E−04	–	–	1.4E−03
	UP 7	–	–	9.6E−04	–	–	1.1E−03
AR	UP 1	7.0E−03	7.0E−03	1.9E−02	1.1E−02	1.1E−02	2.9E−02
	UP 2	9.3E−04	9.3E−04	2.0E−03	1.4E−03	1.4E−03	3.0E−03
	UP 3	8.5E−05	8.0E−05	3.6E−04	4.4E−05	4.2E−05	1.9E−04
	UP 4	1.9E−03	2.1E−05	1.2E−04	3.2E−03	3.2E−05	1.1E−04
	UP 5	1.0E−03	1.0E−03	–	6.6E−04	6.6E−04	–
	UP 6	–	–	1.4E−03	–	–	2.2E−03
	UP 7	–	–	1.5E−03	–	–	1.6E−03

ARP = acid rain precursor and GOP: ground level ozone precursor. BCL = Battelle Columbus Laboratory, GTI = Gas Technology Institute, P I: pyrolysis I pathway (includes UP 1–UP 4 and UP 6), and P II: pyrolysis II pathway (includes UP 1–UP 4 and UP 7). UP 1: unit process 1 (biomass production), UP 2: unit process 2 (biomass transportation), UP 3: unit process 3 (plant construction, decommissioning, and disposal), UP 4: unit process 4 (plant operation and maintenance), UP 5: unit process 5 (hydrogen transport by pipeline), UP 6: unit process 6 (bio-oil transport by truck), and UP 7: unit process 7 (bio-oil transport by pipeline). AR = agricultural residue, FR = forest residue, and WF = whole forest.

Table 6
Key sensitivities and their results.[a]

Case		Path	Base case value EI	GHG	ARP	GOP	Variation from base case (%) EI	GHG	ARP	GOP
1	AR	BCL	57	3.77	1.09E−02	1.65E−02	−43	−45	−48	−50
		GTI	41	2.94	9.03E−03	1.33E−02	−57	−58	−57	−63
		P I	93	6.56	2.24E−02	3.49E−02	−67	−68	−61	−63
		P II	111	8.12	2.25E−02	3.43E−02	−56	−55	−61	−65
2	FR	BCL	43.2	2.96	6.23E−03	8.56E−03	–	−10	−3	−2
		GTI	15.4	1.20	3.72E−03	4.69E−03	–	−8	−2	−2
		P I	21.8	1.58	6.48E−03	9.58E−03	–	–	–	–
		P II	38.9	3.01	6.55E−03	9.01E−03	–	−11	−4	−3
	WF	BCL	49.9	3.77	7.83E−03	1.07E−02	–	−9	−3	−2
		GTI	28.1	2.49	5.68E−03	7.24E−03	–	−10	−3	−2
		P I	15.6	1.43	5.11E−03	7.42E−03	–	–	–	–
		P II	27.5	2.44	5.16E−03	7.02E−03	–	−10	−4	−3
	AR	BCL	57	3.77	1.09E−02	1.65E−02	–	−3	−1	−1
		GTI	41	2.94	9.03E−03	1.33E−02	–	−3	−1	−1
		P I	93	6.56	2.24E−02	3.49E−02	–	–	–	–
		P II	111	8.12	2.25E−02	3.43E−02	–	−5	0	0
3	WF	BCL	49.9	3.77	7.83E−03	1.07E−02	−4	−12	−14	−13
		GTI	28.1	2.49	5.68E−03	7.24E−03	−7	−18	−19	−20
		P I	15.6	1.43	5.11E−03	7.42E−03	−14	−32	−21	−19
		P II	27.5	2.44	5.16E−03	7.02E−03	−8	−18	−21	−20
4	FR	P I	21.8	1.58	6.48E−03	9.58E−03	+103	+87	+13	+45
		P II	38.9	3.01	6.55E−03	9.01E−03	+58	+46	+13	+48
	WF	P I	15.6	1.43	5.11E−03	7.42E−03	+144	+96	+17	+58
		P II	27.5	2.44	5.16E−03	7.02E−03	+82	+56	+17	+61
	AR	P I	93	6.56	2.24E−02	3.49E−02	+24	+21	+4	+12
		P II	111	8.12	2.25E−02	3.43E−02	+21	+17	+4	+12

Case		Path	EI	GHG	ARP	GOP	H	L	H	L	H	L	H	L
5	AR	BCL	57	3.77	1.09E−02	1.65E−02	−5.2	+5	−5.1	+5	−5.6	+5.5	−6	+6
		GTI	41	2.94	9.03E−03	1.33E−02	−7.6	+7.6	−6.3	+6.2	−5.6	+5.6	−6.5	+6.5
		P I	93	6.56	2.24E−02	3.49E−02	−7.5	+7.3	−7.4	+7.3	−6.8	+6.7	−7.2	+7.1
		P II	111	8.12	2.25E−02	3.43E−02	−6	+6	−6	+6	−7	+7	−7.4	+7.3
6	AR	BCL	57	3.77	1.09E−02	1.65E−02	−5.3	+5.8	−5.1	+6.7	−5.6	+7.4	−6	+7.6
		GTI	41	2.94	9.03E−03	1.33E−02	−7.7	+7.9	−6.4	+8.8	−5.6	+7.6	−6.4	+7.9
		P I	93	6.56	2.24E−02	3.49E−02	−8	+10.2	−8	+10.2	−11	+10	−11	+9.8
		P II	111	8.12	2.25E−02	3.43E−02	−6	+9	−6.1	+8.3	−7	+9.8	−7.5	+9.9

BCL = Battelle Columbus Laboratory, GTI = Gas Technology Institute, P I: pyrolysis I pathway (includes UP 1–UP 4 and UP 6), and P II: pyrolysis II pathway (includes UP 1–UP 4 and UP 7). UP 1: unit process 1 (biomass production), UP 2: unit process 2 (biomass transportation), UP 3: unit process 3 (plant construction, decommissioning, and disposal), UP 4: unit process 4 (plant operation and maintenance), UP 5: unit process 5 (hydrogen transport by pipeline), UP 6: unit process 6 (bio-oil transport by truck), and UP 7: unit process 7 (bio-oil transport by pipeline). AR = agricultural residue, FR = forest residue, and WF = whole forest

[a] H and L corresponds to higher and lower value of the sensitivity parameter, respectively. EI stands for energy intensity (MJ/kg H_2), GHG, ARP, and GOP column indicates greenhouse gas intensity (kg CO2 eq/kg H_2), acid rain precursor intensity (kg SO2 eq/kg H_2), and ground level ozone precursor intensity (kg [NO_x + VOC]/kg H_2) respectively.

consuming from the existing cogeneration facility of the upgrading plant. The impact from Case 5 is very significant for all the pyrolysis pathways (Table 6) because bio-oil reforming consumes substantial amounts of natural gas and electricity. The effect of a 10% (Case 5) increase or decrease in biomass yield was also analyzed. This case was developed for all three feedstocks, however, variations in impact were found to be within ±3% of the base case for the WF and FR pathways; hence, only the results from AR are presented in Table 6. Case 6 considers two scenarios; one for an increasing operating factor for the plants (0.7 for year 1, 0.8 for year 2 and 0.95 from year 3 onwards) and the other for a decreasing factor (0.65 for year 1, 0.7 for year 2 and 0.75 from year 3 onwards). In this case too, the variations from the base case were found to be insignificant (±4%) for FR and WF feedstocks, thus only results for AR are presented in Table 6.

4. Conclusion

Among the BCL pathways, the maximum NER (3.33) is that found for FR-based hydrogen; this is followed by the NERs for the WF (2.9) and AR (2.5) pathways, respectively. As with BCL, for the GTI pathways the maximum NER (9.3) is obtained using FR. For P I and P II, the highest NERs are produced using WF; these are 9.1 and 5.2, respectively. Among the BCL and GTI pathways, the lowest GHGs, ARPs, and GOPs are determined for FR-based pathway. For P I and P II, the lowest GHGs, ARPs, and GOPs are for WF-based pathway. NERs for biohydrogen are always higher than the fossil-fuel-based hydrogen, hence, using biohydrogen can improve overall energy efficiency and environmental performance.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.biortech.2011.06.093.

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