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Environmental and economic aspects of hydrogen production and utilization in fuel cell vehicles

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Abstract

A smooth transition from gasoline-powered internal combustion engine vehicles to ecologically clean hydrogen fuel cell vehicles depends on the process used for hydrogen production. Three technologies for hydrogen production are considered here: traditional hydrogen production via natural gas reforming, and the use of two renewable technologies (wind and solar electricity generation) to produce hydrogen via water electrolysis. It is shown that a decrease of environmental impact (air pollution and greenhouse gas emissions) as a result of hydrogen implementation as a fuel is accompanied by a decline in the economic efficiency (as measured by capital investments effectiveness). A mathematical procedure is proposed to obtain numerical estimates of environmental and economic criteria interactions in the form of sustainability indexes. On the basis of the obtained sustainability indexes, it is concluded that hydrogen production from wind energy via electrolysis is more advantageous for mitigating greenhouse gas emissions and traditional natural gas reforming is more favorable for reducing air pollution.

Keywords: Environment; Energy; Sustainability; Hydrogen; Fuel cell; Vehicle

1. Introduction

Modern transportation systems powered by hydrocarbons include increasing numbers of vehicles and are characterized by growing air pollution and greenhouse gas emissions. Such air pollutants as nitrogen oxides (NO_x), volatile organic compounds (VOCs) and carbon monoxide (CO) often are mainly attributable to vehicles [1]. The effects of air pollutants are significant. Nitrogen oxides in combination with VOCs cause the formation of ground level ozone and smog. Healthy people can suffer eye irritation and a decrease in lung function when exposed to smog. NO_x and VOCs react in the presence of sunlight to produce ozone. Elevated levels of ozone can cause lung and respiratory disorders and noticeable leaf damage in many crops, plants and trees. CO emissions impact the ability of red blood cells to transport oxygen to body tissues [2]. Numerous other environmental impacts are associated with emissions of NO_x, VOCs and CO [3,4]. Table 1 presents, for these airborne pollutants, impact weighting coefficients (relative to NO_x) which were obtained by the Australian Environment Protection Authority [5] using cost-benefit analyses of health effects.

The use of hydrogen in fuel cell vehicles can lead to ecologically benign transportation, depending on the source of the hydrogen. Some emissions are associated with the different technologies for hydrogen production [6–8]. Every production process can be characterized by economic, environmental, and other indicators. To evaluate these indicators with one number and to compare a given technology with competitive ones, sustainability indexes were introduced as a sum of normalized economic and environmental criteria with appropriate weighting coefficients. Afgan and Carvalho [9] have considered the hydrogen fuel cell, the natural gas turbine, and photovoltaic and wind energy systems, and evaluated these systems with a multi-criteria method

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Nomenclature

AP	air pollution
CH2	cost of hydrogen
CNG	cost of natural gas
C_nH_m	hydrocarbon
EEQ	energy equivalent, MJ
E	intensity of energy flows, MJ s^{-1}
ΔE	intensity of energy consumption to perform a technology, MJ s^{-1}
EMB	energy embodied in construction materials, MJ
e^-	electron
EOP	operation energy, MJ
H_2	hydrogen
H^+	hydrogen cation
i, j	indexes
IEE	intensity of the energy equivalent, US$ MJ^{-1}
IPM	investments to produce construction materials, US$
LFT	lifetime of unit, s
LHV	lower heating value, kJ
M	mass of the air pollutant, g MJ^{-1}
Q	heat released, kJ
SI	sustainability index
VOC	volatile organic compound
w	weighting coefficient of an air pollutant
W	capacity of hydrogen energy production, MJ s^{-1}
X_{H_2}	fraction of hydrogen

Greek symbols

α	ratio of prices
γ	capital investments effectiveness
η	energy efficiency

Subscripts

AP	air pollution
dir	direct
GHG	greenhouse gas
g	gasoline
H_2	hydrogen
in	input
ind	indirect
ng	natural gas
out	output
oil	crude oil

including the following: performance indicator, market indicator, environment indicator, and social indicator. The indicators are composed of a number of sub-indicators. The evaluation of options under consideration is performed under constraints expressing non-numerical relations among the indicators, and when priority is given to a single indicator and other indicators have the same value.

Table 1
Weighting coefficients of airborne pollutants from motor vehicles

Pollutant	Weighting coefficient
CO	0.017
NO_x	1
VOCs	0.64

Here, a mathematical procedure is proposed to obtain numerical estimates of environmental (air pollution and greenhouse gas emissions) and economic (capital investments effectiveness) criteria, in the form of sustainability indexes. This information is used to evaluate the changes that result from hydrogen implementation as a fuel in fuel cell vehicles, for various hydrogen production technologies.

2. Analysis

Three technologies for hydrogen production are considered: traditional hydrogen production via natural gas reforming, and the use of two renewable technologies (wind and solar electricity generation) to produce hydrogen through the electrolysis of water. The gasoline used in conventional transportation is considered to be obtained by standard crude oil processing and distillation. Economic and environmental indicators are introduced to compare different technologies for gasoline and hydrogen production and utilization in vehicles.

2.1. Economic indicator

Hydrogen and gasoline represent the final products of several modern technologies. The conventional production methods are reforming of natural gas (for hydrogen) and crude oil distillation (for gasoline). The principal technological steps in crude oil and natural gas utilization in transportation are presented in Fig. 1 (left part). Many consider it attractive to generate hydrogen using environmentally benign renewable technologies. The principal technological steps in the utilization of solar and wind energy to produce hydrogen are presented in Fig. 1 (right part).

The overall technology and each technological step in Fig. 1 are accompanied by a consumption of fossil fuel energy ΔE which can be expressed as

$$\Delta E = \Delta E_{\text{dir}} + \Delta E_{\text{ind}} \tag{1}$$

where ΔE_{dir} is the fossil fuel energy directly used to perform a technological step, ΔE_{ind} is the fossil fuel energy used in construction materials and apparatuses, as well as for installation, operation, maintenance, decommissioning, etc. In this paper, fuel energy represents the heat released by the complete burning of all fuel components to CO_2 and H_2O (steam). Thus, the fuel's lower heating value (LHV) is considered. It should be acknowledged that construction materials are also produced from mineral sources (ores, limestone, etc.) which,

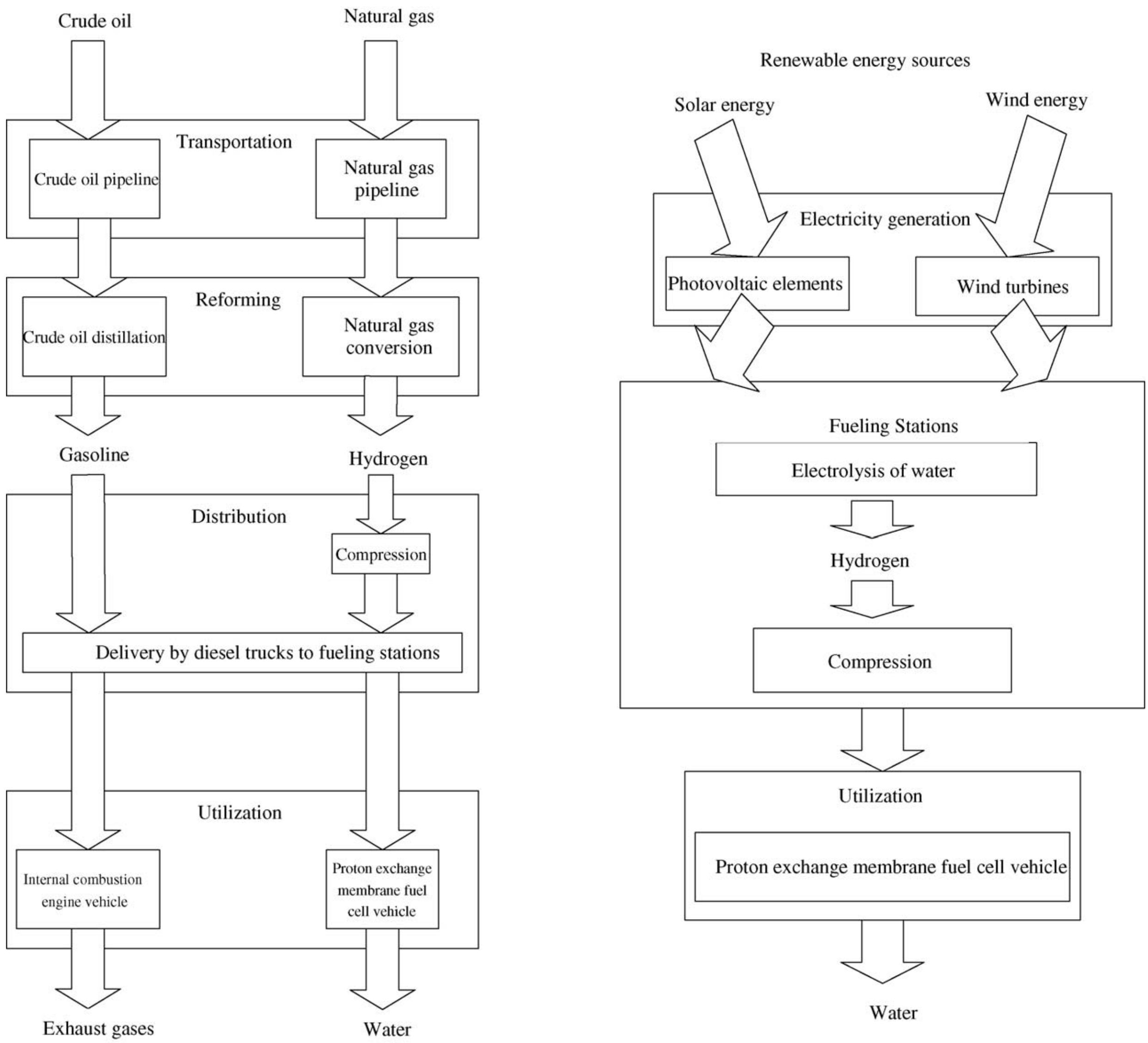

Fig. 1. Principal technological steps in utilizing fossil fuels, and solar and wind energies in transportation.

like fossil fuels, have value. To account for this, the energy equivalent of construction materials and devices (EEQ) is introduced and the following procedure proposed to calculate indirect energy ΔE_{ind}.

The intensities of embodied energies (cost of construction materials and devices per consumed fossil fuel energy to produce them) (IEE) are obtained using Economic Input–Output Life Cycle Assessment Software [10], where they are expressed in US$ 1992 per unit of energy:

$$\text{IEE} = \frac{\text{IPM}}{\text{EMB}} \quad (2)$$

where IPM denotes investments to produce construction materials or devices and EMB denotes energy embodied in construction materials and devices. The fossil fuel energy equivalents of construction materials and devices (EEQ) are calculated as follows:

$$\text{EEQ} = \frac{\text{IEE} \times \text{EMB}}{\text{CNG}} = \frac{\text{IPM}}{\text{CNG}} \quad (3)$$

where the values of embodied energies EMB are taken from [11] and CNG is the industrial cost of natural gas, equal in US$ 1992–0.00316 MJ^{-1} (this year is chosen in order to be consistent with the data obtained by using Life Cycle Assessment Software [10]). The indirect energy to perform a technological operation is evaluated with the following expression:

$$\Delta E_{\text{ind}} = \frac{\sum \text{EEQ} + \text{EOP}}{\text{LFT}} \quad (4)$$

where $\sum$EEQ is the summation of the energy equivalents of construction materials and devices related to a given technological operation, EOP is the operation energy, i.e., the fossil fuel energy required for installation, construction, operation, maintenance, decommissioning, etc. of the equipment, and LFT is lifetime of the unit performing a technological operation.

The fossil fuel and renewable technologies for hydrogen production are generally distinguished by (1) source of energy consumed, (2) efficiency of hydrogen production per unit of energy consumed, and (3) capital investments made per unit of hydrogen produced. To account for all of these differences a new indicator, the capital investments effectiveness γ, is introduced as a measure of economic efficiency. This indicator is proportional to the relationship between the gain and investments and is equal to

$$\gamma = \frac{W(\alpha - 1/\eta)}{\Delta E_{\text{ind}}} \quad (5)$$

The numerator of the fraction in Eq. (5) is proportional to the gain from the exploitation of a technology and the denominator to the investments made in it. Here, α is the ratio in prices of hydrogen (CH2) and natural gas (CNG):

$$\alpha = \alpha_{\text{H}_2} = \frac{\text{CH2}}{\text{CNG}} \quad (6)$$

Also, W is the capacity of hydrogen production expressed in units of hydrogen LHV per second, ΔE_{ind}, the indirect energy which is proportional to the capital investments in a technology, and η is the energy efficiency which is expressible as

$$\eta = \eta_{\text{H}_2} = \frac{E_{\text{H}_2}}{E_{\text{ng}} + \Delta E} \quad (7)$$

In Eq. (7), $E_{\text{H}_2}$is the energy content of hydrogen, E_{ng}, the energy content of natural gas, which is transformed directly to hydrogen, and ΔE is the fossil fuel energy consumption (direct and indirect). Note that $E_{\text{ng}} = E_{\text{H}_2}$ for natural gas reforming and $E_{\text{ng}} = 0$ for "renewable" hydrogen. The initial solar and wind energies do not have any price and, therefore, are not included in the denominator of Eq. (7) for renewable technologies. As a result, the values of η_{H_2} for these technologies can exceed 1.

The same expression (5) can be applied for gasoline production from crude oil, where W is the capacity of gasoline production expressed in units of gasoline LHV per second, α, the ratio in prices of gasoline and crude oil, ΔE_{ind}, the indirect energy of this technology, and η is the energy efficiency of gasoline production from crude oil which is expressible as

$$\eta = \eta_{\text{g}} = \frac{E_{\text{g}}}{E_{\text{oil}} + \Delta E} \quad (8)$$

In Eq. (8), E_{g} is the energy content of gasoline, E_{oil} is the energy content of crude oil, which is transformed directly to gasoline $E_{\text{oil}} = E_{\text{g}}$, and ΔE is the fossil fuel energy consumption (direct and indirect).

The direct and indirect fossil fuel energy consumptions lead to different kinds of pollution. In mass terms greenhouse gases such as CO_2, CH_4, and NO_2 constitute the greatest part of them. The global warming potentials of these substances, according to the International Panel on Climate Change, are 1, 23, and 310, respectively.

To obtain capital investment effectivenesses γ for different technologies it is necessary to consider the steps involved in the transformation of the initial energy sources to the final fuels.

2.1.1. Natural gas and crude oil transportation

These two fossil fuel technologies (Fig. 1) are characterized by their pipeline transportation efficiencies. To evaluate and compare the energy consumption and environmental impact of natural gas and crude oil pipeline transportation, equal lengths of pipelines (1000 km) have been considered. Mechanical work or electricity required to perform pipeline transportation is assumed produced by a gas turbine unit with an average efficiency $\eta_{\text{gt}} = 0.33$ [12].

2.1.2. Natural gas reforming and crude oil distillation

The energy losses in the process of the natural gas reforming comprise approximately 14% of the total energy of the initial flows of methane [13]. The energy efficiency and environmental impact to produce 1 MJ of gasoline have been estimated according to the energy consumption and carbon dioxide emission data of all petroleum refineries in the U.S. in 1996 [14]. The overall results for the direct energy calculations in the reforming and transportation stages are presented in Table 2.

The indirect energy evaluation for the natural gas reforming process is based on the data given by Spath and Mann [6]. In Table 3, the material requirements of the natural gas reforming plant are presented. The values of the energy embodied in the materials and greenhouse gas emissions which accompany this kind of energy consumption have been taken from [11]. In Tables 4a and 4b the indirect energy

Table 2
Total intensity of direct energy consumption and direct greenhouse gas emissions for natural gas and crude oil transportation and reforming (distillation) processes per MJ s^{-1} of fuels produced

Fuel	ΔE_{dir}, MJ s^{-1}	CO_2-equivalent emission, g s^{-1}	η, %
Hydrogen	0.273	75.7	78.5
Gasoline	0.173	12.1	85.3

Table 3
Hydrogen plant material requirements (base case), assuming a 20-year lifetime and 1.5 million Nm^3 day^{-1} hydrogen production capacity (hydrogen LHV production 187.562 MJ s^{-1})

Material	Amount required, metric tonnes	Embodied energy, GJ $tonnes^{-1}$
Concrete	10242	1.4
Steel	3272	34.4
Aluminum	27	201.4
Iron	40	23.5

Table 4a
Energy equivalents and greenhouse gas emissions for a natural gas reforming plant

Materials in natural gas reforming plant	Embodied energy consumption per second of lifetime, MJ s^{-1}	Embodied energy intensity, US$ 1992 MJ^{-1}	Energy equivalent per second of lifetime, MJ s^{-1}	CO_2-equivalent emission, g s^{-1}
Concrete	0.0227	0.0483	0.347	8.44
Steel	0.178	0.0201	1.135	12.82
Aluminum	0.00862	0.0205	0.0559	0.569
Iron	0.00149	0.0201	0.00948	0.107
Total			1.548	21.94

Table 4b
Indirect energy flows and indirect greenhouse gas emissions for the natural gas transportation and reforming stages

Natural gas reforming and transportation processes (energy of natural gas flow: 217.9 MJ s^{-1} based on LHV)	Energy for installation, operation, etc., MJ s^{-1}	CO_2-equivalent emission, g s^{-1}	Indirect energy ΔE_{ind}, MJ s^{-1}	Total CO_2-equivalent emission, g s^{-1}
Natural gas reforming	0.211	21.9	1.759	43.8
Natural gas transportation			2.833	18.7
Total			4.59	62.5
1 MJ s^{-1} of hydrogen			0.024	0.333

calculations are presented. It has been assumed that the operation energy to install, maintain and operate the equipment is equal to the embodied energy which is consumed to produce it. Comparing the values of direct and indirect energies and their emissions, it can be observed that indirect energy (ΔE_{ind}) is more than ten times less than direct one (ΔE_{dir}) and the indirect greenhouse gas emissions are only 0.5% of the direct ones.

No data are available to calculate the indirect energy consumption for the crude oil refinery process. However, as seen in [15], the capital cost of crude oil distillation is lower than that for natural gas reforming. As in the case of the natural gas reforming, the indirect energy use and indirect greenhouse gas emissions are negligible compared to direct ones.

2.1.3. *Renewable hydrogen production via wind energy*

The energy of wind is converted to mechanical work by wind turbines and then transformed by an alternator to AC electricity which is transmitted to the power grid (Fig. 1). The efficiency of wind turbines depends on the location. Applications of wind energy normally make sense only in areas with high wind activity. The data for a 6 MW wind power generation plant was taken from [16] for use here. Tables 5 and 6a,b present the material requirements, indirect energy consumption and indirect greenhouse gas emissions

Table 5
Material requirements and corresponding greenhouse gas emissions for a 6 MW wind power generation plant

Material	Amount required, metric tonnes	Embodied energy, GJ $tonnes^{-1}$	CO_2-equivalent emission, kg $tonnes^{-1}$ of material
Concrete	7647.3	1.4	520
Copper	5.275	131	7450
Fiberglass	496.6	13	804
Steel-carbon/low alloy	1888.0	34.4	2471
Steel-stainless	226.2	53	3280

Table 6a
Indirect energy and greenhouse gas emissions for materials of wind power generation plant, for capacity of 6 MW and lifetime of 25 years

Material	Embodied energy consumption per second of lifetime, MJ s^{-1}	Embodied energy intensity, US$ 1992 MJ^{-1}	Energy equivalent per second of lifetime, MJ s^{-1}	CO_2-equivalent emission per second of lifetime, g s^{-1}
Concrete	0.0136	0.0483	0.208	5.044
Copper	0.000876	0.0412	0.0114	0.0498
Fiberglass	0.00819	0.045	0.117	0.506
Steel-carbon/low alloy	0.0824	0.0201	0.524	5.922
Steel-stainless	0.0152	0.0201	0.0967	0.940
Total			0.956	12.462

Table 6b
Total indirect energy and greenhouse gas emissions for the wind power generation plant

Energy for installation, operation, etc., MJ s^{-1}	CO_2-equivalent emission, g s^{-1}	Total indirect energy, MJ s^{-1}	Total CO_2-equivalent emission, g s^{-1}
0.131	13.60	1.087	26.06

Table 7a
Indirect energy flows and indirect greenhouse gas emissions for hydrogen production via a wind power plant and water electrolysis

Steps to produce hydrogen utilizing wind energy	ΔE_{ind}, MJ s^{-1}	CO_2-equivalent emission, g s^{-1}
6 MW wind power generation plant	1.087	26.06
Electrolysis	0.072	1.47
Total	1.159	27.5

Table 7b
Indirect energy flows and indirect greenhouse gas emissions for hydrogen production via a wind power plant and water electrolysis per MJ s^{-1} of hydrogen produced

ΔE_{ind}, MJ s^{-1}	CO_2-equivalent emission, g s^{-1}	η (Efficiency of fossil fuel energy consumption)
0.289	6.85	3.46

for this plant. Based on data of Spath and Mann [7] for electrolysis to produce hydrogen with a 72% efficiency (on an LHV basis), the indirect energy and greenhouse gas emissions are 6.61 and 5.64%, respectively, of those for a wind power generation plant. Accounting for 7% electricity loss during transmission, the efficiency of hydrogen production is 66.9%. Thus, a 6 MW wind power plant combined with electrolysis of water could produce 4.01 MJ s^{-1} of hydrogen.

Tables 7a and 7b presents the values of indirect energy use and indirect greenhouse gas emissions for hydrogen production via a wind power plant and water electrolysis. The efficiency of fossil fuel energy consumption η reaches 3.46, meaning that the consumed fossil fuel energy (embodied in materials, equipment, etc.) is 3.46 less that the energy of hydrogen produced. The magnitude of η more than 1 is because the energy of wind is considered “free” and is not included into the expression for η. This value should not be confused with the energy efficiency of wind power generation plants, which is about 12–25% and usually calculated as the ratio of electricity produced to the sum of all sources of input energy (mainly kinetic energy of wind).

2.1.4. *Renewable hydrogen production via solar energy*

The indirect energy consumption and indirect greenhouse gas emissions of a photovoltaic system have been evaluated. The photovoltaic elements convert solar energy into direct current (DC) electricity, which is transformed by inverters to alternating current (AC) electricity and transmitted to the power grid. At fuelling stations, ac electricity is used to electrolyze water to produce hydrogen (Fig. 1). Data for a 1.231 kW building-integrated photovoltaic system in Silverthorne, Colorado [17], which utilizes thin film amorphous silicon technology, is considered here. Tables 8a,b,c and 9a,b present the material requirements,

Table 8a
Indirect energy flows for solar cell unit of 1231 kW thin film photovoltaic system, with 157.2 m^2 of surface area and a lifetime of 30 years

Material	Embodied energy, MJ m^{-2}	Embodied energy intensity, US$ 1992 MJ^{-1}	Energy equivalent, MJ m^{-2}	Embodied energy in manufacturing, MJ m^{-2}	Embodied energy in solar cell block, GJ unit^{-1}
Encapsulation	0.2119×10^3	0.0498	3.338×10^3	0.1372×10^3	546.27
Substrate	0.0256×10^3	0.0201	0.163×10^3	0.0564×10^3	34.46
Deposition materials	0.0188×10^3	0.0498	0.296×10^3	0.0925×10^3	61.09
Busbar	0.0051×10^3	0.0498	0.0803×10^3	0	12.63
Back reflector	0.0007×10^3	0.0498	0.011×10^3	0.074×10^3	13.37
Grid	–	–	–	0.0342×10^3	5.38
Conductive oxide	–	–	–	0.0969×10^3	15.23
Total					688.44

Table 8b
Indirect energy flows and greenhouse gas emissions for the units of 1231 kW thin film photovoltaic system

Unit	Embodied energy, MJ unit^{-1}	Embodied energy intensity, US$ 1992 MJ^{-1}	Energy equivalent, GJ unit^{-1}	CO_2-equivalent emission, kg unit^{-1}	CO_2-equivalent emission, g s^{-1}
Inverters	40×10^3	0.0880	111.4	290	0.000306
Wiring	2.9×10^3	0.0509	46.7	180	0.000190
Solar cell block			688.4	7320	0.00774
Total			846.5	7790	0.00823

Table 8c
Total indirect energy flows and greenhouse gas emissions for 1231 kW thin film photovoltaic system

Energy for installation, operation, etc., J s^{-1}	CO_2-equivalent emission per second of lifetime, g s^{-1}	Energy equivalent per second of lifetime, J s^{-1}	Total indirect energy per second of lifetime, J s^{-1}	Total CO_2-equivalent emission per second of lifetime, g s^{-1}
78.957	0.00498	894.81	973.77	0.0132

Table 9a
Indirect energy flows and indirect greenhouse gas emissions for hydrogen production via solar energy utilization and water electrolysis

Steps to produce hydrogen utilizing solar energy	ΔE_{ind}, J s^{-1}	CO_2-equivalent emission, g s^{-1}
1.231 kW photovoltaic system	973.7	0.0132
Electrolysis	64.4	0.0007
Total	1038.1	0.0139

Table 9b
Indirect energy flows and indirect greenhouse gas emissions for hydrogen production via solar energy utilization and water electrolysis per MJ s^{-1} of hydrogen produced

ΔE_{ind}, J s^{-1}	CO_2-equivalent emission, g s^{-1}	η (Efficiency of fossil fuel energy consumption)
1261×10^3	16.88	0.79

indirect energy consumption and indirect greenhouse gas emissions for hydrogen production through photovoltaic power generation and water electrolysis. A procedure similar to that used for the wind power plant is applied to evaluate the indirect energy flows and greenhouse gas emissions associated with electrolysis. Taking into account the efficiency of electrolysis and transmission losses, the 1.231 kW photovoltaic system combined with water electrolysis can produce 823.5 J s^{-1} of hydrogen energy.

2.1.5. Hydrogen compression

The density of hydrogen at standard conditions is low. To assist in storage and utilization as a fuel, the density is increased via compression. The direct fossil fuel (natural gas) energy consumption ΔE_{dir} to compress isothermally 1 mol of hydrogen can be expressed, assuming ideal gas behavior, as

$$\Delta E_{dir} = \frac{R \times T_0}{\eta_{cmp} \times \eta_{gt}} \ln\left(\frac{p_{max}}{p_{min}}\right) \tag{9}$$

Here, $T_0 = 298$ K is the standard environmental temperature and $R = 8.314$ J mol^{-1} K^{-1} is the universal gas constant. Also, η_{cmp} denotes isothermal compression efficiency and η_{gt} gas turbine power plant efficiency.

The direct energy consumed and direct greenhouse gas emissions to compress hydrogen are evaluated, assuming an isothermal compression efficiency η_{cmp} of 0.65 and a typical gas turbine power plant efficiency η_{gt} of 0.33. A typical maximum pressure $p_{max} = 200$ atm [18] is considered. Minimum pressures before compression of $p_{min} = 1$ atm and $p_{min} = 20$ atm are taken for hydrogen production through electrolysis and natural gas reforming [6], respectively.

2.1.6. Hydrogen and gasoline distribution

Hydrogen distribution is replaced by electricity distribution in cases using wind and solar energy (see Fig. 1) and such distribution has been accounted for in hydrogen production. The distribution of compressed hydrogen after its production via natural gas reforming is similar to that for liquid gasoline, but compressed hydrogen is characterized by a lower volumetric energy capacity and higher material requirements for a hydrogen tank.

Table 10
Parameters for capital investments effectiveness γ for different hydrogen production technologies, including hydrogen compression and distribution

Technology	$W/\Delta E_{ind}$	η_{H_2}
Hydrogen from natural gas	26.3	0.70
Hydrogen from wind energy	3.21	1.85[a]
Hydrogen from solar energy	0.78	0.66

[a] This value should not be confused with the energy efficiency of wind power generation plants, which is about 12–25% and usually calculated as the ratio of electricity produced to the sum of all sources of input energy (mainly kinetic energy of wind).

According to the 1997 Vehicle Inventory and Use Survey, the average "heavy–heavy" truck in the U.S. ran 6.1 miles per gallon of diesel fuel [19]. The direct fuel (diesel) energy consumption, assuming a distance of 300 km is traveled before refueling for a truck with a 50 m^3 tank, has been evaluated. The direct energy consumption and greenhouse gas emissions are associated with the combustion of the diesel fuel.

As shown by Lange and Tijm [15], the capital cost of gasoline production from crude oil and hydrogen and synthetic fuel from natural gas will be closer if the capacities of natural gas reforming plants are increased. In 1999, comparing to 1992, the price of natural gas did not change significantly (US$ 0.00318 MJ^{-1}), and Padro and Putsche [20] considered the price of hydrogen produced from natural gas, as US$ 0.005–0.008 MJ^{-1}. Since the ratio in prices of gasoline and crude oil and of hydrogen and natural gas ($\alpha_{H_2} \approx \alpha_g \approx 2$) are also similar [20,21], we assume that the effectiveness of capital investments for gasoline production is equal to that for hydrogen from natural gas for the plants with high capacities, i.e.,

$$\gamma_g = \gamma_{ng} \tag{10}$$

However, to use hydrogen as a fuel for proton exchange membrane fuel cell (PEMFC) vehicles, a power intensive stage for hydrogen compression is required. Therefore, $W/\Delta E_{ind}$ and η are recalculated assuming that the increase in indirect energy (ΔE_{ind}) for compression is proportional to the energy of the fuels directly combusted (ΔE_{dir}) to compress the hydrogen. A factor of proportionality has been estimated through the data for the natural gas transportation and reforming stages (Tables 2 and 4b) where ΔE_{dir} is directly connected with the natural gas compression and combustion processes. Table 10 shows these recalculated values accounting for hydrogen production, compression and distribution.

2.2. Ecological indicators and environmental impacts of PEMFC and ICE vehicles

Ecological indicators characterize the environmental impacts of technologies. Here, the implementation of hydrogen as a fuel for PEMFC vehicles is considered from the point of view of changes in air pollution and greenhouse gas emissions. In a PEMFC stack, electricity (which is converted into mechanical work in electrical motors with efficiency higher than 90%) is generated via the following electrochemical reactions:

$$\begin{aligned} \text{Anode}: 2H_2 &\rightarrow 4H^+ + 4e^- \\ \text{Cathode}: O_2 + 4H^+ + 4e^- &\rightarrow 2H_2O \end{aligned} \tag{11}$$

These reactions occur at low temperature (<100 °C) and involve separation of oxygen from air at the cathode. At these conditions the formation of harmful nitrogen oxides is inhibited and only water is produced during power generation. Thus, the utilization of hydrogen in PEMFC vehicles can be considered as ecologically benign, regarding direct vehicle emissions. Any associated emissions of pollutants and greenhouse gases are associated with hydrogen production.

In ICE vehicles, gasoline (a mixture of hydrocarbons) is combusted in air:

$$C_nH_m + (n + m/4)\,O_2 \rightarrow nCO_2 + m/2\,H_2O + Q \tag{12}$$

The heat Q released during this exothermic reaction is in part converted to mechanical work. According to the Carnot principle, the higher is the temperature of fuel combustion the more mechanical work can be extracted theoretically. The average temperature of the combusting mixture of gasoline and air is about 1300 °C. At such high temperatures the formation of nitrogen oxides is promoted.

Evaporation of gasoline and incomplete combustion lead to emissions of volatile organic compounds and carbon monoxide. Air pollution and greenhouse gas emissions are associated with gasoline production and its utilization in ICE vehicles. Table 11 lists emissions of greenhouse gases and air pollutants during the life cycle from production of hydrogen and gasoline to their utilization in vehicles. The quantities of air pollutants have been evaluated assuming that direct and indirect greenhouse gas emissions (during the production of construction materials and equipment) are from natural gas combustion for all life cycle stages, excluding crude oil distillation. In the latter case, air pollution is evaluated assuming propane combustion. For the stage of gasoline and hydrogen distribution by trucks, air pollution is estimated assuming diesel fuel combustion. The last line in Table 11 represents the emissions which are obtained by using GREET 1.6 software [22]. They are consistent with our data for ICE vehicles powered by gasoline. But an application this model for PEMFC vehicle powered by hydrogen, leads to unsatisfactory results, because in this model the efficiency of a fuel cell vehicle was taken about three times higher compared to the efficiency of ICE vehicle, and energy consumption and emissions connected to the gaseous hydrogen compression were not taken into account.

3. Results and discussion

To characterize air pollution with one measure, the masses m_i of air pollutants in Table 11 are multiplied by the weight coefficients w_i in Table 1 and summarized to obtain the generalized indicator of air pollution AP_j for the entire life cycle of the fuels (gasoline from crude oil, hydrogen from natural

Table 11
Greenhouse gas emissions and air pollution during the life cycle of hydrogen and gasoline production and utilization (per MJ of LHV)

	GHG, g	CO, g	NO_x, g	VOCs[a], g
Hydrogen from natural gas				
Natural gas pipeline transportation and reforming to produce hydrogen	75.7	0.0217	0.0259	0.0544
Hydrogen compression	5.94	0.00413	0.00492	0.000383
Hydrogen delivery to fuelling stations	3.13	0.00722	0.0453	0.00135
Total	84.8	0.0330	0.0761	0.0561
Hydrogen from wind energy				
Electricity generation and hydrogen production via electrolysis at fueling stations	6.85	0.00468	0.00558	0.000435
Hydrogen compression	13.7	0.00949	0.0113	0.000881
Total	20.55	0.0142	0.0169	0.00132
Hydrogen from solar energy				
Electricity generation and hydrogen production via electrolysis at fuelling stations	16.9	0.0115	0.01375	0.00107
Hydrogen compression	13.7	0.00949	0.0113	0.000881
Total	30.6	0.0210	0.0251	0.00195
Gasoline from crude oil				
Crude oil pipeline transportation and distillation to produce gasoline	12.1	0.0120	0.0610	0.0237
Gasoline delivery to fuelling stations	0.19	0.00044	0.00276	0.0000826
Gasoline utilization in ICE vehicle[b]	71.7	0.864	0.0508	0.146
Total	84.0	0.876	0.115	0.170
GREET 1.6 (total)	75.3	0.827	0.072	0.139

[a] Methane emissions are included in VOCs.
[b] Source: Walwijk et al. [24].

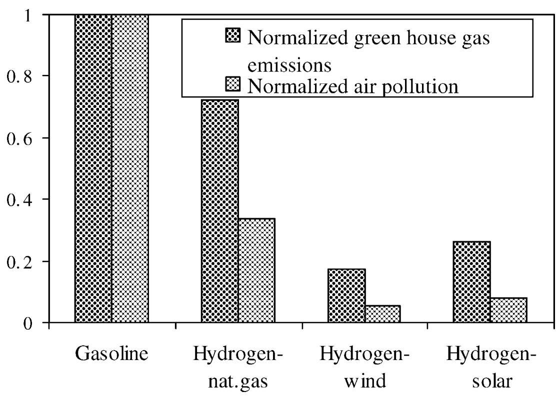

Fig. 2. Normalized emissions of greenhouse gases (GHG) and air pollution (AP) for gasoline and hydrogen use as a fuel for ICE and PEMFC vehicles, respectively.

gas, wind, and solar energies):

$$AP_j = \sum_{1}^{3} m_i \times w_i \tag{13}$$

To find numerical estimates of environmental and economic criteria interaction, the obtained air pollution values and greenhouse gas emissions for gasoline production and different technologies for hydrogen production are normalized to dimensionless values. The normalization procedure involves dividing quantity values by the maximum value for the quantity. Here, we take into account the different average efficiencies (mechanical work per LHV of fuels) of an ICE (0.25) [12] and a PEMFC fuel cell vehicles (0.35) (efficiencies of a fuel cell stack and electrical energy conversion into mechanical work are about 0.4 and 0.9, respectively [23]). After normalizing air pollution and greenhouse gas emissions, the ratios corresponding to one unit of mechanical work production in ICE and PEMFC vehicles are obtained (Fig. 2).

The economic indicator, capital investments effectiveness, depends on α (ratio of prices of hydrogen to natural gas). Using Eq. (10) for gasoline and the values in Table 10, the normalized capital investments effectivenesses at $\alpha_{H_2} = 2$ (ratio of hydrogen to natural gas prices; as it was mentioned in [20] feedstock (natural gas) can account 52–68% of the total cost of hydrogen production for large plants and around 40% for smaller ones) are calculated and presented in Fig. 3. Economic and environmental criteria depend on X_{H_2}, the fraction of hydrogen LHV in the overall energy content of hydrogen and gasoline used as fuels for automobiles. For hydrogen use as a fuel for PEMFC vehicles, accounting for the capital investments effectiveness to produce gasoline and hydrogen, air pollution and greenhouse gas emissions can be expressed as follows:

$$\begin{aligned} \gamma &= X_{H_2} \times \gamma_{H_2} + (1 - X_{H_2})\gamma_g \\ AP &= X_{H_2} AP_{H_2} + (1 - X_{H_2}) AP_g \\ GHG &= X_{H_2} GHG_{H_2} + (1 - X_{H_2}) GHG_g \end{aligned} \tag{14}$$

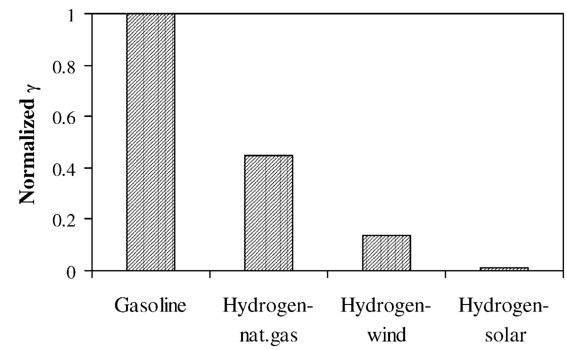

Fig. 3. Normalized capital investments effectiveness γ for gasoline and hydrogen use as a fuel for ICE and PEMFC vehicles, respectively.

where γ, AP, and GHG are normalized capital investments effectiveness, air pollution, and greenhouse gas emissions. Similarly, γ_{H_2}, AP_{H_2}, and GHG_{H_2} are the same quantities for different technologies for hydrogen production, while γ_g, AP_g, and GHG_g are the same quantities for gasoline production.

Fig. 4 presents the normalized capital investments effectiveness γ for different technologies for hydrogen production as a function of the change of the fraction of hydrogen LHV in the overall energy content of hydrogen and gasoline. This effectiveness decreases with the transition from gasoline to hydrogen for any production technologies.

Figs. 5 and 6 present the normalized emissions of air pollution AP and greenhouse gas GHG for different technologies for hydrogen production, as a function of the change in the fraction of hydrogen. These emissions decrease with the transition from gasoline to hydrogen for any production technologies. The first derivative $d\gamma/dX_{H_2}$ corresponds to the intensity of the economic criterion decline with hydrogen fraction. A negative sign implies a decrease in the capital investments effectiveness. The first derivatives dAP/dX_{H_2}, $dGHG/dX_{H_2}$ taken with the opposite sign indicate the intensities of air pollution and greenhouse gas emissions reductions, respectively. The calculated intensities are given in Table 12.

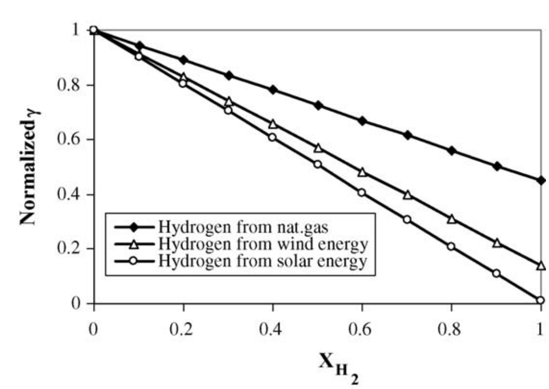

Fig. 4. Normalized capital investments effectiveness γ for several hydrogen production technologies vs. the change of the fraction of hydrogen LHV in the overall energy content of hydrogen and gasoline.

Table 12
Calculated intensities of different technologies for hydrogen production and its use in PEMFC vehicles

Hydrogen production technology	Intensity of decrease in capital investments effectiveness (γ)	Intensity of reduction in air pollution (AP)	Intensity of reduction in greenhouse gas (GHG) emissions
Hydrogen–natural gas	−0.550	0.663	0.279
Hydrogen–wind	−0.861	0.946	0.825
Hydrogen–solar	−0.989	0.920	0.739

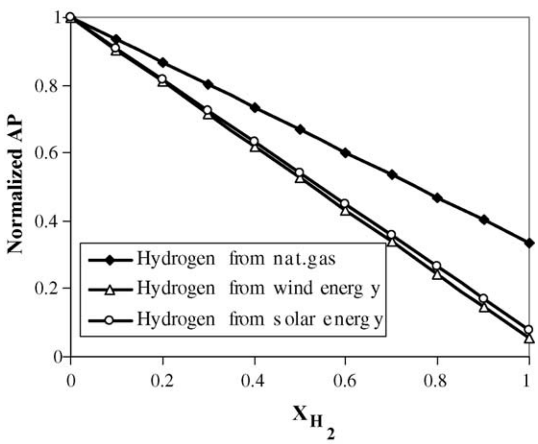

Fig. 5. Normalized emissions of air pollution (AP) for several technologies for hydrogen production and distribution vs. the change of the fraction of hydrogen LHV in the overall energy content of hydrogen and gasoline.

To account for an interaction between environmental and economic criteria as a result of a new technologies implementation, sustainability indexes are introduced. They are obtained by the summation of the corresponding economic and environmental intensities. Thus, the sustainability index for air pollution reduction is the sum:

$$SI_{AP} = \frac{d\gamma}{dX_{H_2}} + \frac{dAP}{dX_{H_2}} \quad (15)$$

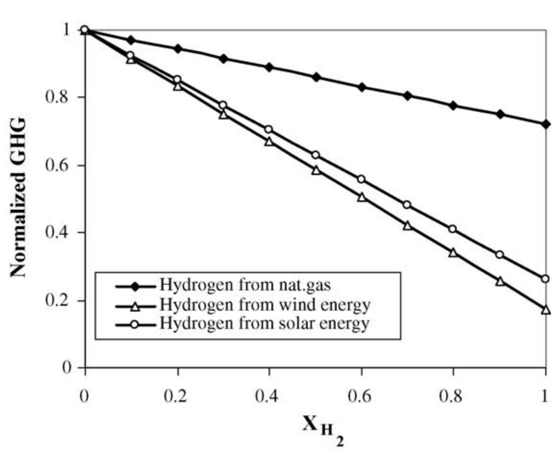

Fig. 6. Normalized emissions of greenhouse gases (GHG) for several hydrogen production technologies vs. the change of the fraction of hydrogen LHV in the overall energy content of hydrogen and gasoline.

Table 13
Sustainability indexes for different technologies for hydrogen production and its use in PEMFC vehicles

Hydrogen production technology	Sustainability index for air pollution reduction	Sustainability index for greenhouse gas emissions reduction
Hydrogen–natural gas	0.113	−0.271
Hydrogen–wind	0.081	−0.036
Hydrogen–solar	−0.069	−0.25

and the sustainability index for greenhouse gas emissions is

$$SI_{GHG} = \frac{d\gamma}{dX_{H_2}} + \frac{dGHG}{dX_{H_2}} \quad (16)$$

The resulting sustainable indexes are presented in Table 13. The negative indexes indicate that the degree of the emissions reduction (environmental criterion) is lower than the degree of the capital investments effectiveness (economic criterion) decline. The positive indexes imply that the reduction of the emissions exceeds the decrease of the capital investments effectiveness. An analysis of sustainable indexes permits to choose an optimal strategy for an implementation of competing environmentally benign technologies. For instance, it can be concluded from the values given in Tables 12 and 13 that the production of hydrogen from wind energy and its application in PEMFC vehicles reduces greenhouse gas emissions more effectively but the traditional technology of natural gas reforming is more favorable for reducing air pollution.

4. Conclusions

Based on life cycle assessments of hydrogen production technologies, a method is proposed to obtain an objective criterion to measure sustainable development, in order to assess hydrogen production technologies for the utilization of hydrogen as an ecologically benign fuel in PEMFC vehicles. The results indicate that a decrease of environmental impact (air pollution and greenhouse gas emissions reduction) as a result of hydrogen implementation as a fuel is accompanied by a decline in the economic efficiency (as measured by capital investments effectiveness). An optimal strategy in an introduction of competing environmentally benign technologies can be chosen taking into account the relationship between environmental and economic criteria in the form of sustainable indexes. Based on the obtained estimations it is concluded that hydrogen production from wind energy via electrolysis is more consistent with sustainable development for greenhouse gas emissions mitigation and traditional natural gas reforming is more favorable for air pollution reduction.

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