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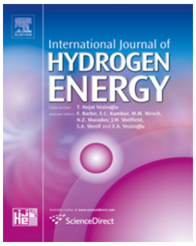

Life cycle greenhouse emissions of compressed natural gas–hydrogen mixtures for transportation in Argentina

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ABSTRACT

We have developed a model to assess the life cycle greenhouse emissions of compressed natural gas–hydrogen (CNG–H_2) mixtures used for transportation in Argentina. The overall fuel life cycle is assessed through a well-to-wheel (WTW) analysis for different hydrogen generation and distribution options. The combustion stage in road vehicles is modeled using the COPERT IV model. Hydrogen generation options include classical steam methane reforming (SMR) and water electrolysis (WE) in central plants and distributed facilities at the refueling stations. Centralized hydrogen generation by electrolysis in nuclear plants as well as the use of solar photovoltaic and wind electricity is also considered. Hydrogen distribution options include gas pipeline and refrigerated truck transportation for liquefied hydrogen. A total number of fifteen fuel pathways are studied; in all the cases the natural gas–hydrogen mixture is made at the refueling station. The use of WE using nuclear or wind electricity appears to be less contaminant that the use of pure CNG.

1. Introduction

In internal combustion engines, hydrogen can be used as pure gas or as a mixture, mainly with compressed natural gas. Successful experience with pure hydrogen has been attained in commercial vehicles, both using fuel cell technology and H_2-fueled internal combustion engine. Presently, the compressed natural gas–hydrogen (CNG–H_2) mixture is being used in California to drive city buses [1]. CNG–H_2 mixtures provide a cleaner fuel than CNG since the CO_2, CO and HC emissions decrease with increasing H_2 content; however NO_x emissions increase [2]. Under certain conditions related to air to fuel ratio, the given power is increased using CNG–H_2 mixtures [3]. Depending on the vehicles fleet replacement, the use of CNG–H_2 mixtures as transport fuel will probably decrease the urban emission burden however it is necessary to consider the environmental burdens with a life cycle point of view. Production and distribution of hydrogen includes processes which emit air pollutants and greenhouse gases (GHG). Depending on the raw material these processes include fossil fuel extraction and transport, electricity generation, gas compression, gas liquefaction, gaseous or liquid transport and dispensing. Different life cycle assessment (LCA) studies has been conducted for H_2 production, Spath and Mann [4] present a comprehensive analysis for natural gas steam reforming (SMR), concluding that >74% of the life cycle GHG emissions come from the H_2 production plant. LCA of H_2 production pathways from natural gas considering gas and liquid H_2 distribution has been conducted by Wang et al. [5], this work also presents results for different hydrogen vehicle fleet penetration. LCA of H_2 production by different pathways including water electrolysis has been conducted by Wietschel

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et al. [6] presenting ten different options including H_2 distribution, concluding that electricity production in the case of water electrolysis and the SMR for natural gas-based production are the most polluting stages.

We have developed a model to assess the life cycle GHG emissions of CNG–H_2 mixtures used for transportation in Argentina. The overall fuel life cycle is assessed through a well-to-wheel analysis for different H_2 generation and distribution options. Since Argentina has an extensive fleet of CNG vehicles and a widespread CNG distribution structure, this way of H_2 utilization is considered as a natural first step for H_2 penetration as transportation fuel. The study covers from raw material extraction to fuel combustion in light duty vehicles; a comparison with pure CNG fueled vehicles is also reported.

2. Life cycle greenhouse emissions quantification

The fuel pathways considered account for the following H_2 generation process: SMR, water electrolysis in central plants and distributed facilities at the refueling station and centralized generation by water electrolysis in nuclear plants. Distribution options include gas pipeline and refrigerated truck transportation for liquefied H_2. As a result fifteen fuel pathways are analyzed. Greenhouse gas emissions of the national electricity generation system are considered to estimate GHG emissions from electricity use of each stage in the fuel pathways considered. The fuel pathways considered are characterized in Table 1 below.

Table 1 – Definition of CNG–H_2 pathways studied.

Fuel pathway acronym	Description
SMRP	H_2 production by Steam Methane Reforming and gaseous distribution through pipeline.
SMRP (Seq.)	Idem SMRP with CO_2 sequestration.
SMRL	H_2 production via Steam Methane reforming and distribution in liquid form.
SMRL (Seq.)	Idem SMRL with CO_2 sequestration
DSMR	Distributed generation of H_2 via Steam methane reforming.
CEP	H_2 production by Water Electrolysis and gaseous distribution through pipeline. Electricity from the National Mix.
CEL	H_2 production via Water Electrolysis and distribution in liquid form. Electricity from the National Mix.
DE	Distributed generation of H_2 via Water Electrolysis. Electricity from the National Mix.
SECP	H_2 production by Water Electrolysis and gaseous distribution through pipeline. Electricity from Solar Photovoltaic.
SECL	H_2 production via Water Electrolysis and distribution in liquid form. Electricity from Solar Photovoltaic.
WECP	H_2 production by Water Electrolysis and gaseous distribution through pipeline. Electricity from Wind generation.
WECL	H_2 production via Water Electrolysis and distribution in liquid form. Electricity from Wind generation.
NEP	H_2 production by Water Electrolysis in a Nuclear Power Plant and gaseous distribution through pipeline.
NEL	H_2 production via Water Electrolysis in a Nuclear Power Plant and distribution in liquid form.
CNG	Compressed Natural Gas pathway.

Two types of feedstock are considered: natural gas and hydrogen. For natural gas, life cycle stages include: extraction, processing, pipeline transport and intermediate compression. The life cycle stages considered for natural gas feedstock are shown in Fig. 1, where E indicates electricity consumption in intermediate compression stages for natural gas transport and GHG indicates the greenhouse gases emissions.

For hydrogen feedstock, life cycle stages depend on the generation step. For H_2 generation by SMR the natural gas feedstock is considered together with the reforming stage and H_2 transport options, Fig. 2 shows the three pathways studied. The dotted line from the SMR process indicates the possibility of carbon dioxide capture and sequestration; this option is possible only for H_2 generation in central plants. GHG emissions from fuel combustion in cryogenic trucks are accounted for in the liquid hydrogen (LH_2) transport stage.

For generation via water electrolysis, the fuel pathways studied are shown in Fig. 3. The fuel pathways considered include the utilization of electricity from the national generation mix and electricity generated in nuclear power plants and from two renewable sources (see Section 2.1). In Argentina, the electricity sector includes thermal, hydro and nuclear power plants. Thermal power is generated using primarily natural gas, complemented by fuel oil, gas oil and coal.

Thermal power generation technologies include steam turbines, gas turbines and combined cycle units representing more than 50% of the total electricity generation. Hydroelectric power generation represents 35–40% of the total generation. Nuclear power generation is carried out in two nuclear power plants, accounting for ~10% of the total generation offer. For the estimation of GHG emissions from the electricity life cycle, the following stages were considered:

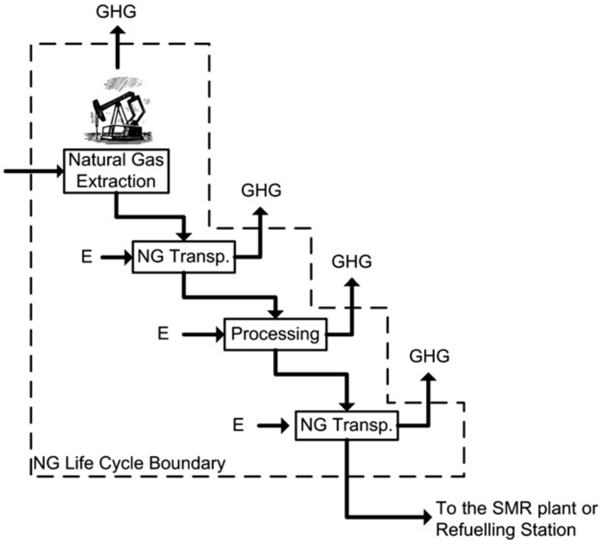

Fig. 1 – Natural gas life cycle.

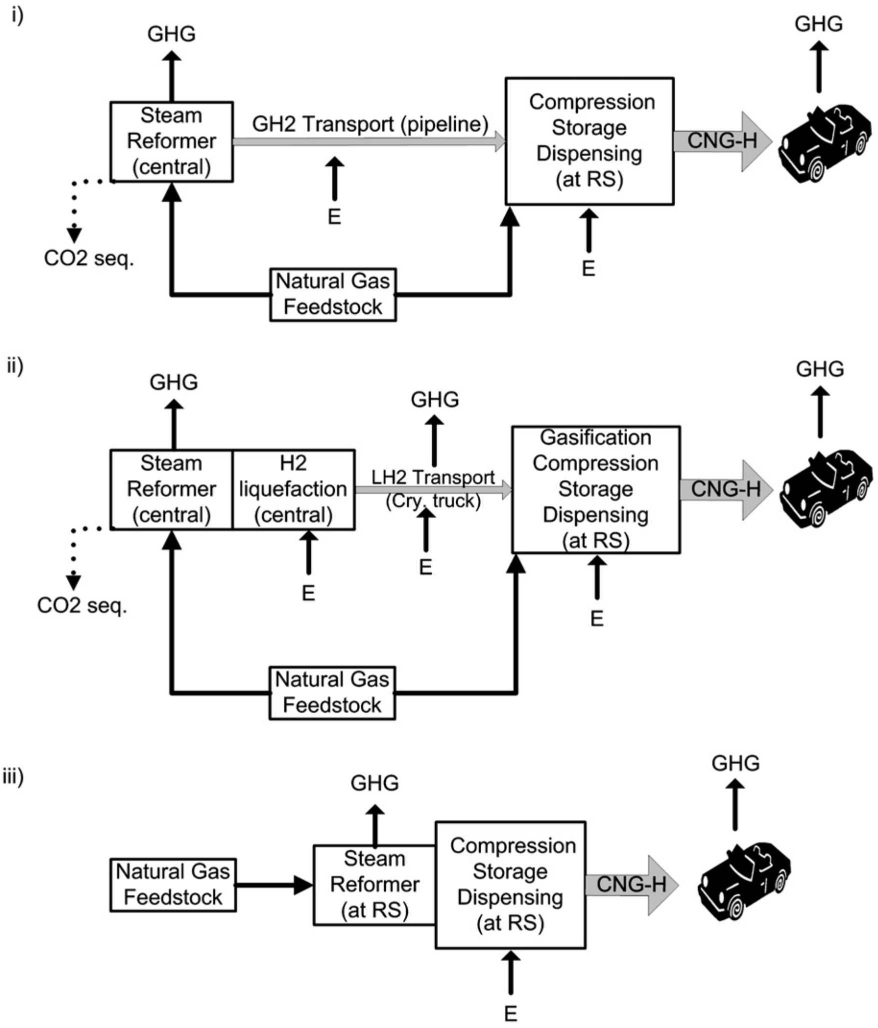

Fig. 2 – Selected fuel pathways for H_2 production via SMR: i) generation in a central plant and distribution through gas pipeline, ii) generation in a central plant and distribution of liquefied hydrogen in refrigerated trucks, iii) Distributed generation of hydrogen by SMR at the refueling station.

extraction and processing of raw materials, transport, refining (where it is applicable) and electricity generation itself. For hydroelectric power generation, the biomass decay during dam operation was considered. For nuclear power generation, emissions of GHG gases are present mainly in the uranium fuel cycle. Further details of the Argentinean electricity life cycle GHG quantification could be found in Martinez and Eliceche [7].

In this work, the three major GHGs specified in the Kyoto protocol namely, CO_2, CH_4 and N_2O, were considered. Emissions from these three gases were combined together with their global warming potentials (1 for CO_2, 21 for CH_4, and 310 for N_2O) to derive CO_2-equivalent GHG emissions.

The mass of each GHG was calculated using the emission factors presented by several literature sources, as summarized below. The 2006 IPCC guidelines [8] were the basis for natural gas fuel cycle, except for transport stage where the National GHG Inventory [9] data was used. Wietschel et al. [6] emission factors were used for H_2 feedstock life cycle assessment, the GHG emission factor for electricity supply was taken from national data [10]. The emission factor for national diesel fuel combustion in cryogenic trucks was corrected and a transport distance of 500 km was considered. Natural gas composition used for the analysis is an average of the major gas production areas in the country.

2.1. *Nuclear and renewable electricity utilization*

A worldwide trend that is gaining relevance in the last few years is to produce hydrogen in nuclear power plants [11]. Such a process uses a huge amount of heat duty to conduct a water thermal cracking often requiring high temperatures (e.g. 850 °C). New types of nuclear reactors are needed to supply this heat requirement, which will be commercially available before 2030 in an optimistic forecast [12]. Until then, we propose to use electricity from nuclear power to conduct traditional water electrolysis. As shown later, this technology option offers the possibility of H_2 production with a very low life cycle GHG emissions. Nuclear power use only gives the possibility of centralized H_2 production. Since the Argentinean

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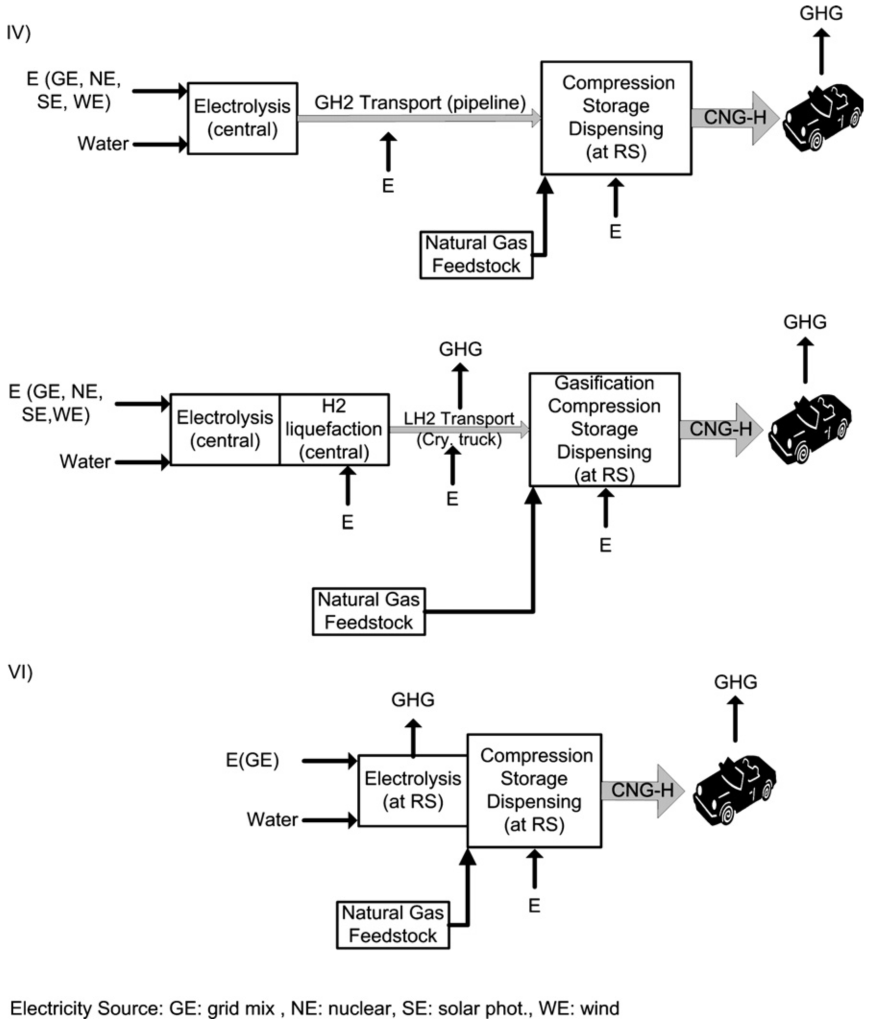

Fig. 3 – Selected fuel pathways for H_2 production via water electrolysis: IV) generation in a central plant and distribution through gas pipeline, V) generation in a central plant and distribution of liquefied hydrogen in refrigerated trucks, VI) Distributed generation of hydrogen by water electrolysis at the refueling station.

electricity grid distributes an undifferentiated mix of the overall electricity produced in the country, it is not technologically possible to use nuclear electricity in a distributed manner. As it was stated in the previous section, life cycle assessment of nuclear electricity includes emissions during the uranium fuel cycle, which is composed by the following stages: uranium mining and milling, conversion, enrichment, fuel fabrication, operation and waste disposal. The GHG emissions during the uranium fuel cycle are generated from fuel combustion in mining and milling stages, and in an indirect way from electricity consumption in conversion, enrichment, fuel fabrication, operation and waste disposal stages [7].

Concerning the use of renewable electricity sources, we propose using solar photovoltaic and wind energy [13,14]. We suggest to produce H_2 in a centralized electrolysis plant near the solar field and wind farms, respectively. The life cycle GHG emissions of solar photovoltaic and wind electricity generation include those arising from the fabrication of the generation unit of each renewable energy type. The solar photovoltaic panel fabrication includes stages like silica extraction, silica transformation and panel assembling. The stages involved in wind turbine fabrication include iron and copper extraction and the production of steel, resins, fiber glass and concrete.

The GHG emission factors for these two renewable electricity sources was adapted from data presented by Evans et al. [15] which include an extensive literature review of the solar photovoltaic and wind turbine fabrication processes considering the life cycle stages detailed above.

2.2. *Greenhouse emissions quantification from fuel combustion*

The combustion step was assessed using COPERT IV [16] model for pure CNG fueled vehicles. The fuel consumption of CNG light duty vehicle considered in the model is 0.08 m^3/km.

Due to lack of experimental data, the CO_2 emissions for CNG–H_2 mixture combustion in road vehicles are modeled based in the carbon content of the local fuels. Similar fuel

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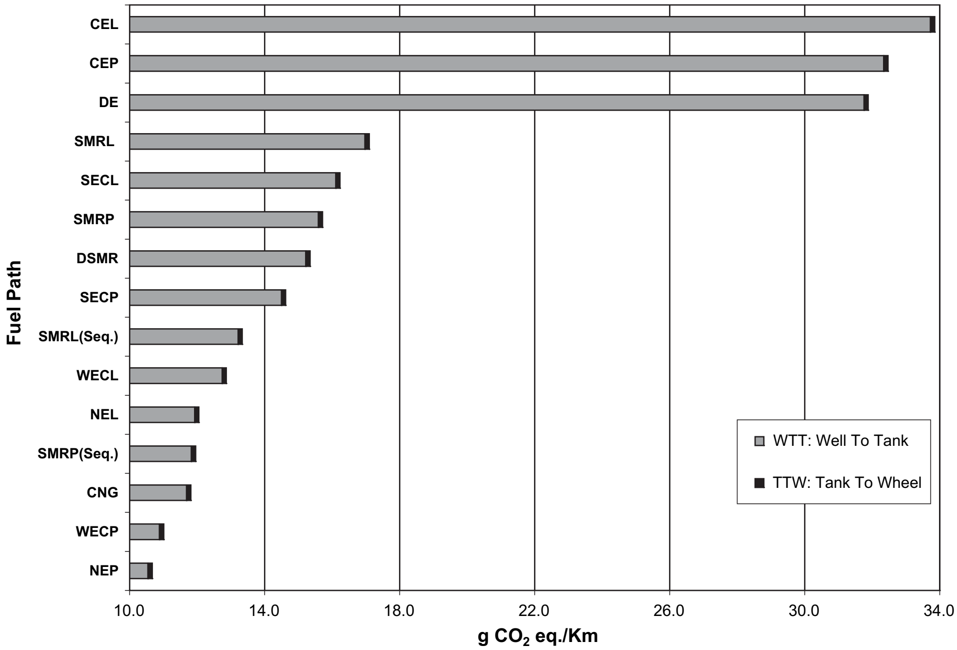

Fig. 4 – Well-to-wheel GHG emissions for CNG–H_2 pathways.

consumption in terms of energy usage per kilometer was considered for the CNG–H_2 mixture vehicle in each fuel pathway option. Other pollutant emissions as CH_4, N_2O, CO and other hydrocarbons will be estimated in future engine operation tests with different hydrogen content in the gas mixture.

Finally, construction stages and pipeline fabrication were not considered for the assessment.

3. Results and discussion

Well-to-wheels analysis is comprised of two components, the ‘well-to-tank’ (all activities involved in producing the fuel), WTT, and ‘tank-to-wheel’ (the operation/driving of the vehicle), TTW [17].

A distance of 1 km city transport had been chosen as the functional unit for the study. Fig. 4 shows the well-to-wheel GHG emission results of the CNG–H_2 fuel pathways analyzed expressed in g CO_2 eq./km of city transport distance traveled by a light duty vehicle. The mixture considered is composed by 90% of CNG and 10% of hydrogen.

4. Conclusions

The life cycle GHG emissions of CNG–H_2 (90–10) mixture were analyzed for fifteen different fuel pathways from primary fuel extraction and transformation to fuel mixture combustion in light duty vehicles. For 12 of the 14 options considered, the life cycle GHG emissions for vehicles using CNG–H_2 mixture are higher than those for pure CNG vehicles (11.82 g CO_2 eq./Km). Compared with pure CNG vehicle, all SMR hydrogen pathways have higher GHG emissions including those with CO_2 sequestration. Hydrogen generation by water electrolysis via electricity from a nuclear power plant or from wind farms together with hydrogen pipeline distribution presents lower GHG emissions than pure CNG pathway and appears to be the most promising options for CNG–H_2 mixture utilization in Argentina in terms of GHG mitigation.

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