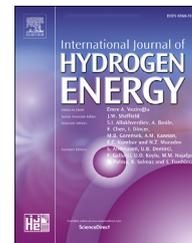




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A techno-economic evaluation of the hydrogen production for energy generation using an ethanol fuel processor

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HIGHLIGHTS

- Techno-economic evaluation of the hydrogen production was performed.
- Fuel processor used monolith catalysts for the ethanol reformer and WGS reactor.
- H₂ production price was competitive with H₂ from refueling stations in California.
- The H₂ production cost was significantly affected by the ethanol price.
- The use of compact fuel processor using monolith catalysts is economically feasible.

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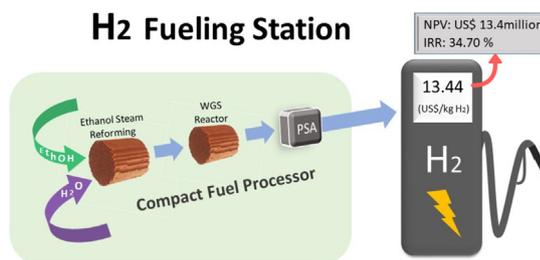
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GRAPHICAL ABSTRACT



ABSTRACT

The techno-economic analysis of a process to convert ethanol into H₂ to be used as a fuel for PEM fuel cells of H₂-powered cars was done. A plant for H₂ production was simulated using experimental results obtained on monolith reactors for ethanol steam reforming and WGS steps. The steam reforming (Rh/CeSiO₂) and WGS (Pt/ZrO₂) monolith catalysts

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remained quite stable during long-term startup/shut down cycles, with no carbon deposition. The H₂ production cost was significantly affected by the ethanol price. The monolith catalyst costs contribution was lower than that of conventional reactors. The H₂ production cost obtained using the expensive Brazilian ethanol price (0.81 US\$/L ethanol) was US\$ 8.87/kg H₂, which is lower than the current market prices (US\$ 13.44/kg H₂) practiced at H₂ refueling stations in California. This result showed that this process is economically feasible to provide H₂ as a fuel for H₂-powered cars at competitive costs in refueling stations.

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Introduction

The growing concern with the reduction of greenhouse gas emissions and atmospheric pollution has increased the interest in using fuel cells as a power source. When compared to combustion engines, fuel cells are considered as more efficient devices for energy production [1]. In this scenario, hydrogen, if produced from renewable sources, can contribute to the sustainable production of energy, since it can be converted electrochemically into energy in a fuel cell, to produce electricity for various applications, such as electronic devices laptops, vehicles and homes [2]. In the case of hydrogen fuel cell electric vehicles (FCEVs), the automotive industry is interested in this technology to minimize emissions; reduce fuel consumption; decrease noise from traffic in cities and increase vehicle efficiency [3–5]. According to the literature [3], between 2030 and 2050, the demand for FCEVs fueled by hydrogen is expected to increase, which requires the establishment of a widespread hydrogen refueling station (HRS) infrastructure [3,4].

The technology for H₂ production is well established for large-scale centralized plants and it is based on hydrocarbon steam reforming [6,7]. The majority of hydrogen is produced in refineries to upgrade crude oil (hydrocracking and hydro-treating process), in the petrochemical industry to synthesize different chemical compounds (such as ammonia and methanol), for oil and fat hydrogenation, metallurgical processes (as a reduction gas), among many other uses [8]. In any case, hydrogen is not produced to be used as a fuel for energy generation. Today, the installed infrastructure for hydrogen production and distribution is not able to meet the demand of the market for its use as an energy vector.

Moreover, the establishment of a new hydrogen production and distribution infrastructure represents an economical and technical obstacle that limits the widespread use of hydrogen as an energy carrier and thus its use in FCEVs [4]. Then, in order to supply hydrogen at low cost, it must be produced locally, using an existing fuel distribution infrastructure. In addition, the use of a renewable source for the production of hydrogen will enable FCEVs to achieve zero emission [3].

Several authors studied the production of hydrogen for HRS using onsite electrolysis integrated with renewable energy sources such as wind and solar [9–11]. Nevertheless, studies about hydrogen production from bioethanol for HRS

were not found in the literature. As bioethanol is a renewable source, its use for on-site hydrogen production in HRS would be very interesting, especially in countries such as Brazil and the United States. In these countries, ethanol is currently mixed with gasoline and then, its production and distribution infrastructure is already available. In addition, ethanol is significantly less toxic than methanol or gasoline.

Hydrogen can be obtained from ethanol by different processes and raw materials such as the steam reforming of bioethanol, a water and ethanol mixture that may be produced by biomass fermentation [12–14].

Finally, unlike industrial plants, on-site H₂ generators must obviously be compact.

However, on site production of H₂ faces a new challenge: the development of small-scale reformers [7]. Large scale hydrogen plants cannot simply be reduced in size to meet the economic and safety aspects required for use in fuel cells and hydrogen refueling stations. For example, lower pressures and temperatures are needed for small-scale reformers to be competitive. Strong limitations of heat and mass transfer occurs on conventional multi-tubular reactors using catalysts in the form of pellets and are not adequate to the dynamics of these new systems. Then, the design of new reactors and catalysts is fundamental for the success of new technologies based on the production of H₂ using biomass-derived liquids.

Most of studies on ethanol steam reforming have been conducted considering various formulations of powdered catalysts that are not useful for industrial or remote applications (small scale H₂ reformers). On the other hand, the monolithic structures can be used as supports to obtain catalytic reactors that have high structural and thermal stability, in order to minimize pressure drop and avoid reactor blocking [15]. The advantages of using monoliths over particulates are: high geometric surface area with enhanced heat and mass transfer; low pressure drop; structural stability of thin wash-coated catalysts; excellent heat management with heat exchanges; rapid response to transient operations; decreased reactor weight and volume; no activation for precious metal catalysts during start up and no special requirements for discharge (exposure to air); precious metals are recycled and reused.

Monolithic structures washcoated with noble metal-containing catalysts have high activities comparing to transition metal catalysts, allowing a decrease in the amount of metal used. Furthermore, the pre-reduction of precious

metals is not required and they do not undergo oxidation to air exposure, being more tolerant to process poisons. They are also more resistant to carbon formation than transition metal catalysts and can be regenerated if coke is deposited [16]. A short review highlighting the advantages of monolithic catalysts for H₂ generation is available in literature [17].

However, the formation of carbon during the conversion reactions of ethanol in monolithic catalysts is noteworthy. Therefore, it is necessary to develop monolithic catalysts that are stable under ethanol conversion reactions and resistant to carbon formation in order to enable the commercialization of this technology. Recently, a new Rh/CeSiO₂ catalyst supported on a ceramic monolith was tested for the production of hydrogen through steam reforming (SR) of ethanol [18]. The stability of the monolith supported catalyst for long-term test was investigated as well as its performance in startup/shutdown cycles and regeneration for coke removal. The monolith supported catalyst remained quite stable during long-term operation without catalyst deactivation.

Although in recent years the number of studies in the literature on steam reforming ethanol has increased [19–27], some technical and economic issues need to be overcome to commercialize the technology. The major obstacle is the cost associated with the fuel processor. Before starting the development of a process and executing the design of a plant, economic evaluations must be conducted. This evaluation will determine whether the project should be undertaken, abandoned or reformulated [28]. Even if the technical information available is not sufficient to design a complete plant, conducting an economic analysis is important and will determine if the project would be economical and financially viable. The economic evaluation of a process passes through several steps: determination of the process flowchart; calculation of mass and energy flows; sizing of the main equipment; capital cost estimate; production cost estimate; forecast of the selling price of the product; and estimation of return on investments [28].

Song and Ozkan [29] developed an economic analysis model based on cost structures in the United States for H₂ production from a bio-ethanol steam reforming process. The process includes feedstock considerations and also hydrogen purification strategies. Two different capacities levels (1500 and 150,000 kg of H₂/day) were analyzed. The estimation of the hydrogen selling price and the contribution of different cost components were evaluated to determine the feasibility of commercialization of a bio-ethanol steam reforming process for hydrogen production. Sensitivity analyses were performed and the effect of utility cost, feed cost, catalyst cost, reaction temperature and GHSV addressed. According to the authors, the price variation of the hydrogen can be mainly attributed to the cost of the feedstock. The authors found that the use of precious metal catalysts requires a H₂ selling price of US\$ 22.34/kg, as opposed to US\$ 4.27/kg required for a transition metal catalyst. However, this analysis assumes that the space velocities and catalyst life time are identical but the proper noble metal catalyst is much more active, stable and requires a considerably lower loading than base metal options, however is more expensive depending on the use of Rh (highly expensive) or Ru (1/10 as expensive as Rh). For this reason, the assumption does not correspond to the reality. The final

selling price of H₂ does not appear to be very sensitive to reactor temperature. However, there are several disadvantages of using high reaction temperatures that were not evaluated by the authors. The use of high temperatures may increase material costs, bring additional safety concerns and since the heat could not be recycled with high efficiency, increased temperatures would increase the utility costs.

Lopes et al. [30] performed a technical and economic analysis of an ethanol reformer prototype developed by Hytron to produce hydrogen to be used in a 5 kW PEMFC. The authors evaluated the hydrogen and electricity cost produced with this hydrogen in a PEMFC for stationary and distributed generation of electricity. The system considered by the authors to supply electricity is composed basically by a fuel processor, a fuel cell and an electricity conditioning system. The fuel processor contains: the ethanol reformer, the reforming gas treatment system and the purification module. For the economic investment analysis, costs with materials replacement (catalysts and process water), equipment, labor, ethanol and electricity were considered. In order to determine the cost of the hydrogen produced by the fuel processor, a flow chart analysis was used. The authors obtained a hydrogen production cost of US\$ 14.11/kg of H₂ using an ethanol price of US\$ 0.371/L. The costs with the equipment of the reformer and ethanol represented 54.0% and 32.9% of the H₂ production price, respectively.

Even though there are detailed analyses of the economics involved in the H₂ generation through steam reforming of ethanol process by fuel processors [29–33], no economic analysis of fuel processors using monolith catalysts for the ethanol reformer and WGS reactors for H₂-powered car refueling stations is available. Therefore, the main goal of this work was to simulate a compact fuel processor using experimental data obtained with monolithic catalysts to be used in H₂-powered car refueling stations and to perform a techno-economic evaluation of this process. With the introduction of fuel cell vehicles on the market [34–36] refueling stations will be needed to supply hydrogen at competitive costs. Because of technical difficulties of storing large amounts of hydrogen, costs of the gas distribution networks and safety aspects associated with this technology, decentralized systems of hydrogen production will be needed in refueling stations. Since ethanol is currently mixed with gasoline in fuel stations in countries such as Brazil and USA, the infrastructure for ethanol production and distribution is already well established. Therefore, the local production of H₂ from bio-ethanol by a compact fuel processor using monolith catalysts is presented as a viable strategy to reduce the sale price of H₂ to support fuel cell powered cars.

Materials and methods

Catalytic monoliths preparation

The preparation of Rh/CeSiO₂ catalyst deposited in cordierite monolith for SR of ethanol was previously described [18]. First, SiO₂ (Aerosil 380) was dissolved in an alkaline solution and mixed with an ammonium hydroxide solution. After addition of this solution to a (NH₄)₂Ce(NO₃)₆·6H₂O (Aldrich 98%)

solution, a precipitate was obtained. Then, the precipitate was dried (383 K) and calcined (773 K for 5 h) under air flow. The support (CeSiO_2) was impregnated with an aqueous solution of $\text{RhCl}_3 \cdot x\text{H}_2\text{O}$ to obtain a catalyst containing 1% wt Rh. After impregnation, the sample was dried (383 K) and calcined (673 K for 5 h) under air flow.

For the preparation of catalytic monolith, a slurry was obtained with 40% catalyst powder in ethanol and then, acetic acid was added. The slurry was ball milled during 3 h to reduce particle size and to provide a good adherent washcoat. Several steps were used to washcoat the monoliths (OD of 18 mm, 19 mm long, cell density of 400 cpsi – provided by BASF). First, one end of the monolith was submerged in the slurry for 5 min and then, it was rotated and dipped into the slurry for another 5 min to provide an homogeneous coating of the walls. For the purpose of ensure the proper washcoat loading, this process was repeated as many times as necessary. To remove the excess slurry from the channels, the coated monoliths were shaken for several minutes and then partially dried in a gentle flow of warm air. The coated monoliths were calcined in muffle at 773 K for 2 h and the final washcoat loading was obtained by measuring the changes in the monoliths weigh before and after the impregnations. The monoliths contained loadings of 0.97 and 0.47 g/in^3 .

A Pt/ZrO₂ catalyst was synthesized for testing on the WGS reaction. For synthesis of the catalyst, a commercial monoclinic ZrO₂ support supplied by Norpro was used. The catalyst containing 1% wt Pt supported on ZrO₂ was obtained by wet impregnation using an aqueous solution of 8% H_2PtCl_6 in H_2O . After impregnation, the sample was dried at 383 K for 12 h and calcined at 673 K for 5 h under air flow. The catalyst was deposited on cordierite monoliths (18 mm in diameter, 19 mm in length, 400 cpsi cell density - BASF) by the washcoat method following the same procedure previously described. Two monoliths were prepared with catalyst loadings of 0.94 and 0.77 g/in^3 .

A thermal shock test similar to that used to insure adhesion of the washcoat on automotive emission control monolith catalysts was done. Initially, the washcoated ceramic monolith was cycled to high temperature (973 K) for 1 h and then quickly exposed to a jet of cold air. After repeated thermal cycles only a 1.2% of mass loss was noted.

Catalytic tests

SR of ethanol using the washcoated monolith was carried out in gas phase at 1 atm and 1023 or 1028 K. The reaction mixture with a total gas flow of 150 mL/min and a water to ethanol molar ratio of 3.5 was obtained by the injection into the reactor using a syringe pump. The cordierite monoliths were immobilized, with Fiberfrax wrapping, in a quartz tube reactor (ID of 20 mm, OD of 26 mm). The reaction products were on-line injected in a gas chromatograph (GC Agilent 3000 Micro A), containing three channels equipped with a thermal conductivity detector (TCD) and three columns: a molecular sieve, a Poraplot Q and OV-1 column.

The WGS reaction was performed at 623 K using a gas mixture with a total gas flow of 50 mL min^{-1} and a composition of 4% CO, 2% CO₂, 14% H₂, 15% H₂O and N₂ (balance) in order to reproduce the outlet of the ethanol fuel reformer conditions. The two monoliths prepared were used simultaneously in the reactor with a distance of approximately 10 cm of each other. Additional water was added into the gas stream after the SR of ethanol reactor by using a saturator at 348 K, which slightly changed composition of the inlet stream to the WGS reactor. Before reaction, the catalysts were reduced under a H₂ (50 mL min^{-1}) from room temperature to 623 K (10 K min^{-1}) for 1 h. The system was then purged with N₂ (50 mL min^{-1}). The reaction was carried out for 570 h of time on stream (TOS) with start/stop mode. The reactor effluent was analyzed by an on-line by gas chromatograph (Micro GC Agilent 3000 A), containing three channels equipped with a thermal conductivity detector (TCD) and three columns: a molecular sieve, a Poraplot Q and OV-1 column.

Description of the hydrogen production process

The simulation of the hydrogen production plant through steam reforming of ethanol was based on the flowchart shown in Fig. 1. It was considered a continuous process of H₂ production with a fuel processor and H₂ storage tanks. The fuel processor was composed of the reformer reactor, the Water Gas-shift (WGS) reactor and the Pressure Swing Adsorption (PSA) unit. The steam reforming of ethanol occurs in the reforming reactor while the purification of hydrogen

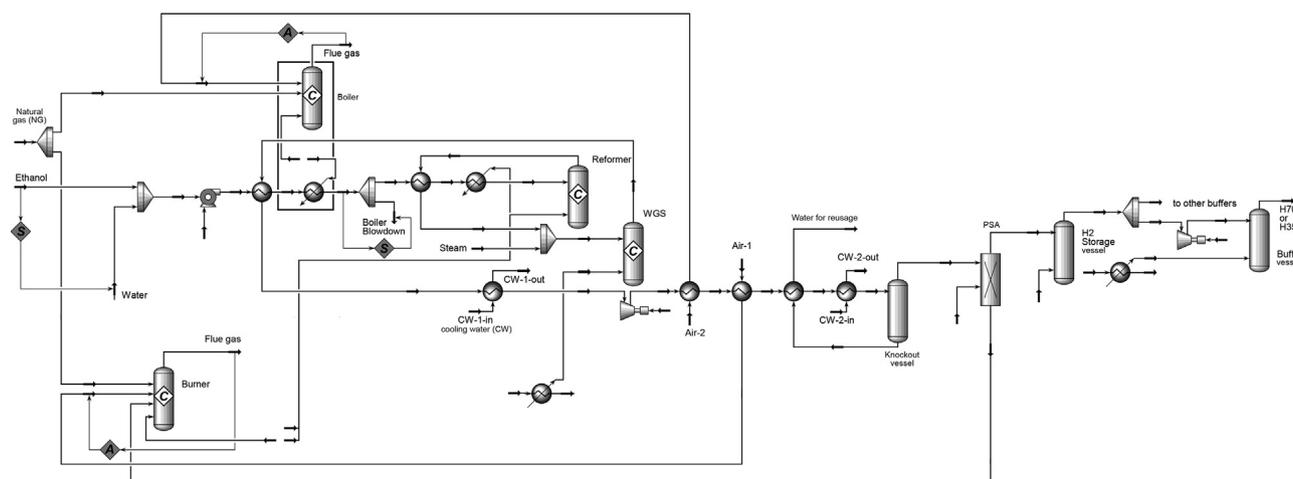


Fig. 1 – Fuel processor unit flowchart for economic analysis.

stream takes place on the WGS reactor and PSA unit. The H₂ stream with high purity was then stored in tanks that will be used to supply hydrogen to fuel cell-powered cars.

Ethanol and steam feed the reformer while natural gas was used as a fuel in the boiler and as fuel in the burner of the reformer. The complete combustion of natural gas was achieved by controlling the flow rates of Air 1 and Air 2 streams. Then, more steam is added to the syngas obtained by the steam reforming of ethanol to meet the molar composition used in the experiments described in results section. This stream was then used as a feed for the WGS reactor, where CO and H₂O are converted to CO₂ and H₂.

The outlet stream of the WGS reactor was compressed to obtain the required operating pressure of the PSA [37] and then cooled in a series of heat exchangers. A knock-out vessel was used to separate the liquid products from the stream. The gas fraction of the stream, after separation of the liquids, goes through the PSA unit for H₂ purification. The PSA waste gas stream containing CO, CO₂, CH₄ and unrecovered H₂ is burned to ensure that only CO₂ and H₂O will be released to atmosphere. The water condensed on the knock-out vessels was sent to a water treatment utility simulated as described by Junqueira et al. [38] to be reused, which contributes to reduce the water consumption cost.

The pure H₂ produced was cooled and sent to a buffer system for compression to 80 MPa as required for refueling stations for fuel cell-powered cars.

The simulation of the key components of the plant and the economic analysis of the process were carried out using operating conditions, assumptions and methodologies described in the next sections.

Ethanol feedstock

The necessary infrastructure for ethanol distribution is already well established in countries like Brazil and the US, since ethanol is currently blended with gasoline so the infrastructure for ethanol is already available at fueling stations. The price of ethanol used in this work was the average ethanol price at the distributor in the city of Rio de Janeiro provided by the Brazilian National Oil Agency in 2018 [39].

Fuel processor

The fuel processor consists of the ethanol reformer, the WGS reactor and the PSA unit and they will be described in more detail next.

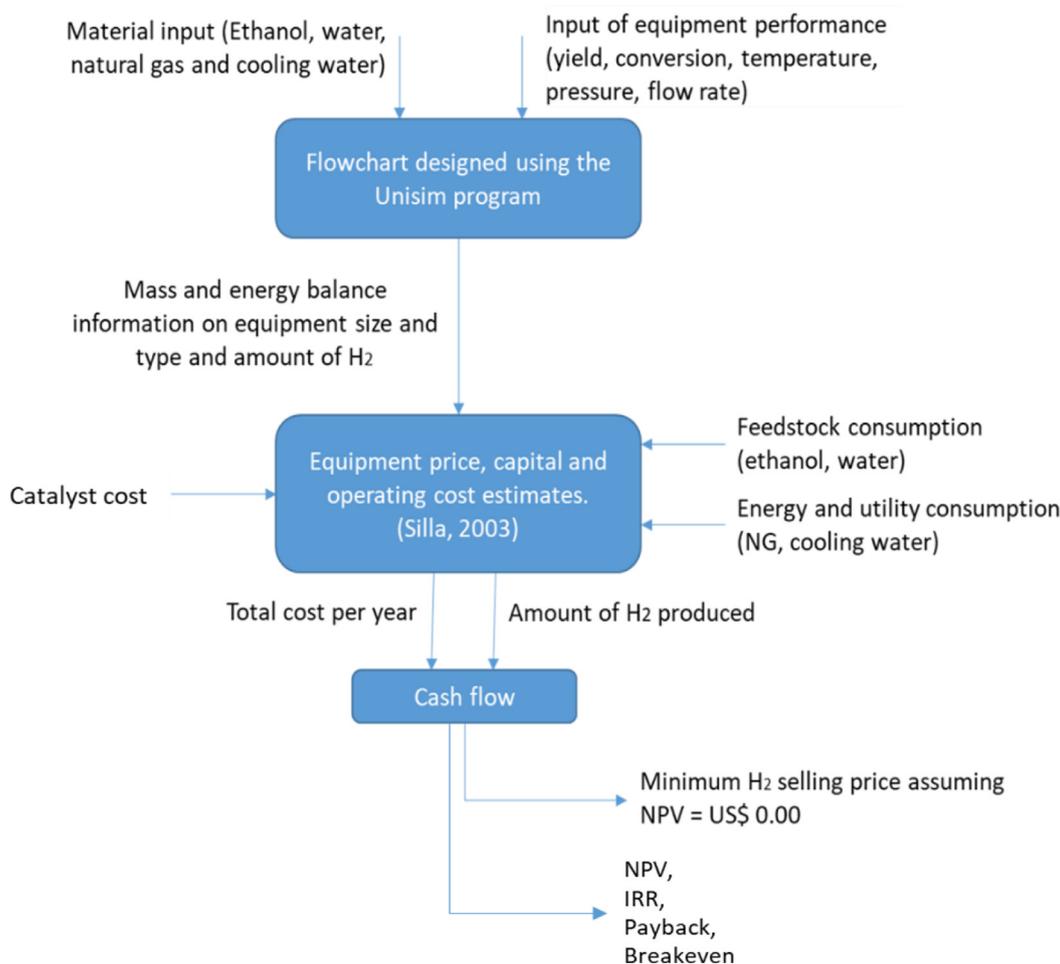
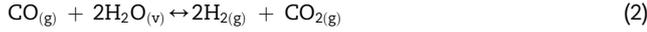
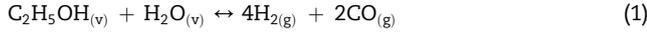


Fig. 2 – The overview of the economic analysis (Adapted from da Silva et al. [27]).

Ethanol reformer

The ethanol reformer reactor was simulated as a conversion reactor. The ethanol and water conversions used to simulate it were the values obtained from experimental results, which will be described in detail in economic analysis section. Ethanol steam reforming (1) and WGS (2) reactions are shown below.



Carbon formation was not considered since our experimental results showed no carbon formation after extensive aging.

The ethanol and water conversions and the molar com-

The ethanol flow rate used (660 L/h) was required to obtain sufficient H_2 production to completely power 600 H_2 -powered cars per day.

WGS reactor and PSA unit

The WGS reactor was also simulated as a conversion reactor. It was considered the conversion of CO and H_2O into CO_2 and H_2 . The conversion of reactants and the molar composition of the products and the unreacted CO and H_2O obtained for WGS were calculated from Eqs. (9)–(14):

$$\text{CO conversion (\%)} = \frac{[\text{CO}]_{\text{in}} - [\text{CO}]_{\text{out}}}{[\text{CO}]_{\text{in}}} \quad (9)$$

$$\text{H}_2\text{O conversion (\%)} = \frac{[\text{H}_2\text{O}]_{\text{in}} - [\text{H}_2\text{O}]_{\text{out}}}{[\text{H}_2\text{O}]_{\text{in}}} \quad (10)$$

$$\text{H}_2 \text{ molar composition (\%)} = \frac{[\text{H}_2]_{\text{out}}}{[\text{H}_2]_{\text{out}} + [\text{CO}]_{\text{out}} + [\text{CO}_2]_{\text{out}} + [\text{H}_2\text{O}]_{\text{out}}} \quad (11)$$

$$\text{CO molar composition (\%)} = \frac{[\text{CO}]_{\text{out}}}{[\text{H}_2]_{\text{out}} + [\text{CO}]_{\text{out}} + [\text{CO}_2]_{\text{out}} + [\text{H}_2\text{O}]_{\text{out}}} \quad (12)$$

$$\text{CO}_2 \text{ molar composition (\%)} = \frac{[\text{CO}_2]_{\text{out}}}{[\text{H}_2]_{\text{out}} + [\text{CO}]_{\text{out}} + [\text{CO}_2]_{\text{out}} + [\text{H}_2\text{O}]_{\text{out}}} \quad (13)$$

$$\text{H}_2\text{O molar composition (\%)} = \frac{[\text{H}_2\text{O}]_{\text{out}}}{[\text{H}_2]_{\text{out}} + [\text{CO}]_{\text{out}} + [\text{CO}_2]_{\text{out}} + [\text{H}_2\text{O}]_{\text{out}}} \quad (14)$$

positions of the products (H_2 , CO , CO_2 and H_2O) were calculated from Eqs. (3)–(8):

$$\text{C}_2\text{H}_5\text{OH conversion (\%)} = \frac{[\text{C}_2\text{H}_5\text{OH}]_{\text{in}} - [\text{C}_2\text{H}_5\text{OH}]_{\text{out}}}{[\text{C}_2\text{H}_5\text{OH}]_{\text{in}}} \quad (3)$$

$$\text{H}_2\text{O conversion (\%)} = \frac{[\text{H}_2\text{O}]_{\text{in}} - [\text{H}_2\text{O}]_{\text{out}}}{[\text{H}_2\text{O}]_{\text{in}}} \quad (4)$$

The PSA unit was considered to operate at 316 K and 1 MPa in order to obtain a H_2 stream with high purity (99.99%) [37].

Economic analysis

The approach used in the development of the process design and economic analysis is shown in Fig. 2. For this analysis, the

$$\text{H}_2 \text{ molar composition (\%)} = \frac{[\text{H}_2]_{\text{out}}}{[\text{H}_2]_{\text{out}} + [\text{CO}]_{\text{out}} + [\text{CO}_2]_{\text{out}} + [\text{H}_2\text{O}]_{\text{out}}} \quad (5)$$

$$\text{CO molar composition (\%)} = \frac{[\text{CO}]_{\text{out}}}{[\text{H}_2]_{\text{out}} + [\text{CO}]_{\text{out}} + [\text{CO}_2]_{\text{out}} + [\text{H}_2\text{O}]_{\text{out}}} \quad (6)$$

$$\text{CO}_2 \text{ molar composition (\%)} = \frac{[\text{CO}_2]_{\text{out}}}{[\text{H}_2]_{\text{out}} + [\text{CO}]_{\text{out}} + [\text{CO}_2]_{\text{out}} + [\text{H}_2\text{O}]_{\text{out}}} \quad (7)$$

$$\text{H}_2\text{O molar composition (\%)} = \frac{[\text{H}_2\text{O}]_{\text{out}}}{[\text{H}_2]_{\text{out}} + [\text{CO}]_{\text{out}} + [\text{CO}_2]_{\text{out}} + [\text{H}_2\text{O}]_{\text{out}}} \quad (8)$$

Table 1 – Values of economic parameters for the NPV evaluation.

Item	Assumption
Plant lifetime	25 y
Plant operation time	333 d/y [20]
Depreciation schedule length	10 y [20]
Ethanol feedstock cost (2018)	US\$ 0.81/m ³ [31]
Debit cost	1,05% [33]
Interest rate	10%
Total tax rate	38%
Debit weight	80% [33]
Term of financing	10 y
Installed capital cost for PSA	US\$ 100,000.00
Operating cost for PSA system	US\$ 0.184/1000 Nm ³ /d [34]
Carbon credits	US\$ 15.00/tCO ₂
Energy supply	US\$ 0.24/kWh
Natural gas	US\$ 0.60/m ³ [35]
Water supply	US\$ 5.64/m ³ [36]

Table 2 – Composition of the inlet and outlet stream of reformer and WGS reactors.

Compounds	Molar composition (%)			
	Ethanol Reformer		WGS Reactor	
	Inlet	Outlet	Inlet	Outlet
Ethanol	22.2	0.0	0.0	0.0
H ₂	–	51.3	39.5	50.1
H ₂ O	77.8	25.5	42.4	29.4
CO	–	15.6	11.0	0.9
CO ₂	–	7.6	7.1	19.6

first step was to develop the flowchart to simulate the plant for H₂ generation. It was used along with literature information and experimental results to simulate a process using the UniSim software. The energy and material balance from the UniSim model were used to size equipment and to determine total capital and operating costs.

Total capital and production costs are required in order to determine the financial attractiveness of the process. According to Silla [28], the total production cost is composed by: direct costs, indirect costs and general costs. Direct costs, also called variable costs, are proportional to production rates, while indirect costs are composed of fixed costs and general plant costs. General costs include costs of managing the firm, marketing, and researching and developing of new and old products [28]. The sum of indirect and general cost was considered as the fixed cost. Table 1 shows all the costs and financial parameters that were taken into account on the calculation of production cost. Since steam reforming and WGS catalysts exhibited high stability and the reforming catalysts can be completely regenerated [18], as it will be discussed in more detail in the Results and Discussion section, the exchange of the catalysts was not taken into account in the economic evaluation. However, if catalyst regeneration is required, this could be carried out during the maintenance that was considered in the economic analysis, since the plant was designed to operate for 333 d/y (Table 1), which corresponds to 8000 h of operating. Furthermore, the debt cost related to amortization during 10 years was already paid. In addition, the resources required for the replacement of the catalysts were foreseen in the depreciation fund in the period of 10 years (Table 1). Since there are no longer amortization debts, it is possible to incur a new investment for the replacement of any equipment.

All costs were determined using information obtained from [28], with the exception of the boiler costs provided by [40], the operating cost of the PSA system obtained from [37], compressors from [41,42]. The price of reforming and WGS catalysts were assumed as 100 times the price of a commercial catalyst (US\$ 10.3/Kg) [29,43].

The net present value (NPV) was estimated using Eq. (15), where t is the year number in the evaluation period; i is the interest rate; C_t is the cash flow on t and C_0 is the initial investment.

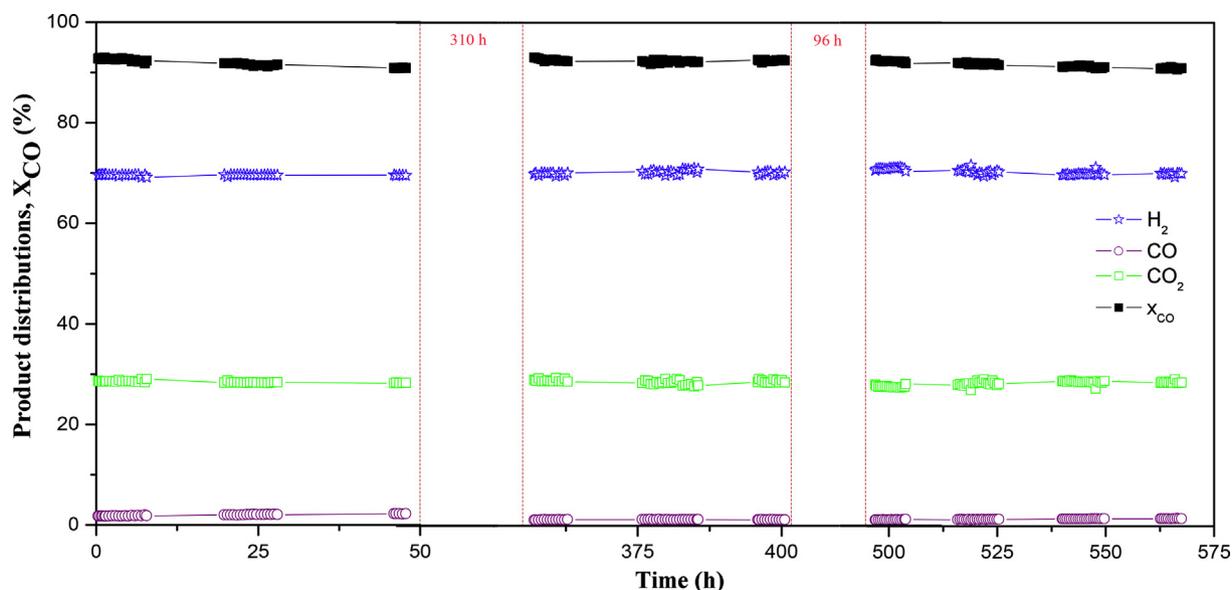


Fig. 3 – CO conversion and product distribution obtained for WGS reaction at 573 K with Pt/ZrO₂ monolith (washcoat loading 0.94 and 0.77 g/in³) on/off cycles.

Table 3 – Simulated data obtained for the composition of the inlet and outlet stream of reformer and WGS reactors.

Compounds	Molar composition (%)			
	Ethanol Reformer		WGS Reactor	
	Inlet	Outlet	Inlet	Outlet
Ethanol	22.2	0.0	0.0	0.0
H ₂	–	54.4	39.8	50.8
H ₂ O	77.8	22.1	43.0	32.0
CO	–	16.2	11.9	0.9
CO ₂	–	7.3	5.3	16.3

$$NPV = \sum_{t=1}^T \frac{C_t}{(1+i)^t} - C_0 \quad (15)$$

Once the capital and operating costs were determined, the information was put into an Excel spreadsheet set up to calculate the minimum hydrogen selling price considering NPV equals to zero.

A NPV was calculated to determine the feasibility of the processes for a hydrogen selling price practiced in the market [44]. The internal rate of return (IRR) was determined for an interest rate that makes the NPV crosses the *i* axis on a NPV versus *i* graphic. While the breakeven is the production rate that makes the revenue and total costs intersect and the payback is the time, in years, necessary to recover the initial investment (capital cost).

Results and Discussion

The experimental and simulated data obtained for the reformer and WGS reactors, and the economic analysis of the process are presented and discussed next.

Catalytic tests

The performance of a Rh/CeSiO₂ catalyst deposited on cordierite monolith for long-term SR of ethanol at 1028 K was investigated [18]. The initial ethanol and water conversions were 100 and 67.2%, respectively, using a space velocity (WHSV) of 4.1 h⁻¹. The catalyst remained stable during 96 h of time on stream (TOS) and the products formed were only H₂, CO and CO₂. CH₄ was not detected, indicating that it was completely reformed (Table 2). Raman spectroscopy did not reveal the deposition of carbon after long-term operation, which is consistent with catalyst stability. The stability of the catalyst was also tested after start/stop conditions to simulate the daily operation of the refueling station. Four startup/shutdown cycles were carried out, in which the SR of ethanol at 1023 K and 8.5 h⁻¹ was performed during 24 h, followed by cooling to 523 K and remaining at this temperature for 3 h. The reactor was then re-heated to 1023 K and the reaction re-started for more 24 h. The catalyst was stable during the four cycles with no carbon formation detected by Raman analysis.

CO conversion and product distributions for WGS reaction at 623 K over Pt/ZrO₂ monoliths were shown in Fig. 3. The same start up/shut down procedure used for SR of ethanol was also investigated for the WGS with a WHSV of 1.9 h⁻¹. The reaction was carried out for 50 h of TOS and then stopped for 310 h and started again up to 40 h. Then the reaction was reinitiated after 96 h and total TOS was 570 h. The CO conversion remained stable around 91.8% and the water conversion was 30.7% during the three cycles, indicating that catalyst does not deactivate during 570 h of TOS. The outlet stream of the WGS reactor contained H₂, CO, CO₂ and H₂O (Table 2).

The fuel processor was simulated using the experimental results shown above: 100% of ethanol conversion in the reformer reactor with a WHSV of 4.1 h⁻¹; and 92% of CO

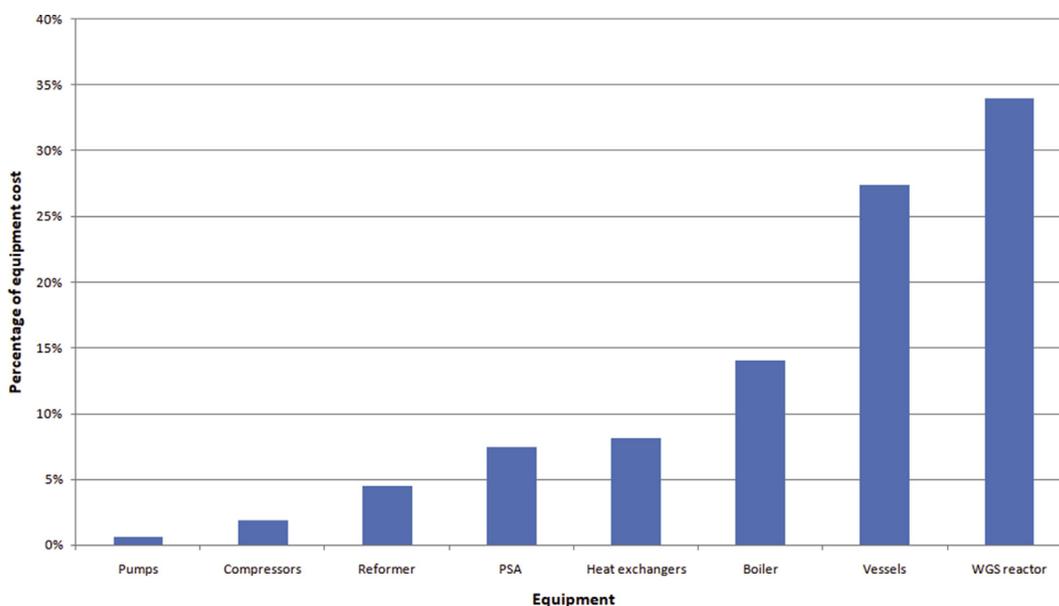


Fig. 4 – The contribution from each type of equipment, in the total equipment cost.

H₂ Production Cost

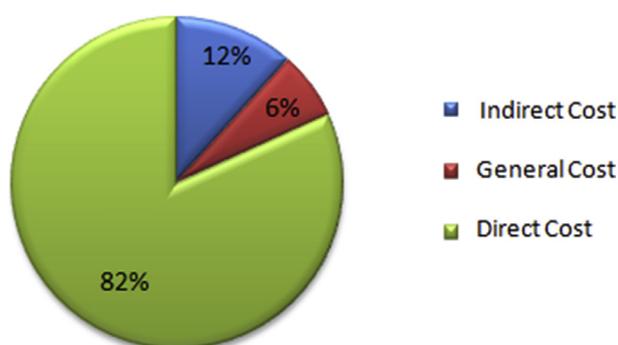


Fig. 5 – Cost of H₂ production and the contribution of different components costs.

conversion in the WGS reactor with a WHSV of 1.9 h⁻¹, producing 2.43 m³ of H₂/L of ethanol fed at 303 K and 101 kPa (1603.26 m³ H₂ produced per 659.06 L ethanol fed).

The simulated data obtained for the composition of the inlet and outlet stream of reformer and WGS reactors (Table 3) shows that the compositions values found were very close to the values obtained experimentally.

Economic evaluation

After designing the flowchart of the plant for H₂ production, the contribution of each type of equipment, in the total equipment cost, was calculated and it is shown in Fig. 4. The WGS reactor contribution was more significant representing approximately 35% of the total equipment cost. WGS reactors are usually expensive due to the large size needed to maintain the reaction temperature and large monolith volume required. Since the temperature gradient along the WGS reactor can be very large and the catalyst can only reach a high

activity in certain temperature ranges, an effective temperature control of the reactor is needed. The costs with vessels include the H₂ storage vessel, knockout vessel, deaerator, ion separation beds, water tanks and filter [38]. The highest contribution is mainly due to the quantity of equipment considered and the special material and pressure factors for the H₂ storage vessel.

For the operational conditions, a hydrogen production cost of US\$ 8.87/kg H₂ was obtained. Fig. 5 shows the cost of H₂ production and the contribution from the different components costs. The contributions from direct costs were more significant, corresponding to 82% of the total cost of hydrogen production.

The cost of H₂ production and the contributions of the main categories related to direct costs are presented in Fig. 6. The costs of feedstock (Fig. 6) and consequently the costs of ethanol contribute significantly to the total cost of hydrogen production mainly due to the highest ethanol price (Fig. 7). These results showed that the cost of hydrogen production was strongly influenced by the price of ethanol for a fuel processor, which produces hydrogen for a refueling station using ethanol as a resource.

The relevance of the contribution of the ethanol price on economic viability of an industrial steam reforming unit for production of hydrogen from ethanol was reported by Roldán [33]. Song et al. [29] also showed that the ethanol price contributes significantly to the selling price of hydrogen produced from steam reforming at two production scenarios in the United States: (i) forecourt production scale (1,500 kg H₂/day) and (ii) central production scale (150,000 kg H₂/day). Moreover, contrary to what has been reported in the literature [29], the results obtained in this work showed that the contribution of the catalyst price for the costs of hydrogen production was very low even though it was used rhodium and platinum noble metal catalysts. This was likely due to the use of smaller amounts of catalysts on monolith reactors compared to conventional reactors (fixed-bed reactors and pellets). In addition, in this work, the catalysts costs represent around 20% of the

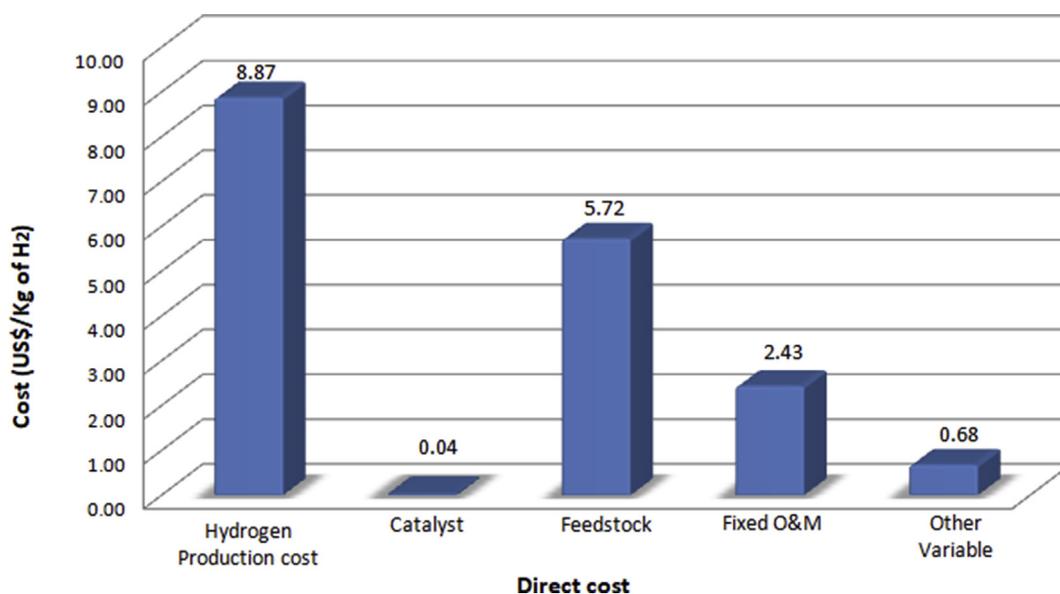


Fig. 6 – The contributions of the main categories related to direct costs on the cost of H₂ production.

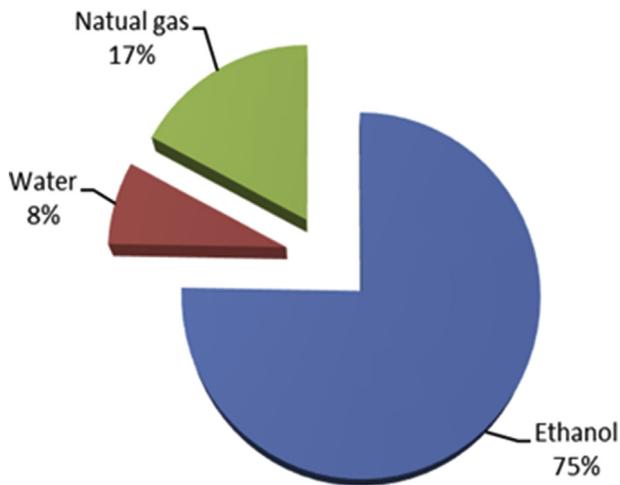


Fig. 7 – Feedstock components cost contribution in the feedstock total cost.

reforming section costs. Therefore, the reforming section costs using monolith reactors are lower than those obtained when conventional reactors are used.

Sensitivity analysis

Since the ethanol price was the main contribution for the final H_2 production cost, a sensitivity analysis of its price on the hydrogen production cost was performed (Fig. 8). The Brazilian ethanol price (0.81 US\$/L ethanol) was used as base value and the sensitivity analysis was done with a variation range of $\pm 40\%$. The results showed that the hydrogen production cost varied from 10.59 to 7.17 US\$/kg H_2 , when the ethanol price was decreased from 1.13 to 0.48 US\$/L ethanol, respectively. Using the E85 (ethanol fuel blend of 85% ethanol fuel and 15% gasoline) market price of 0.74 US\$/L ethanol reported in the literature for California fuel stations [45], the H_2 production cost obtained was 8.52 US\$/kg H_2 .

A sensitivity analysis of plant lifetime of hydrogen production cost was also carried out (Fig. 9). A variation range of

$\pm 60\%$ was considered, using a plant lifetime of 25 years as base value (US\$ 8.87/kg H_2). Increasing the plant lifetime from 10 to 40 years, hydrogen production cost decreased from 9.59 to 8.69 US\$/kg, respectively.

Taking into account the need of catalyst exchange during 25 years of plant operation, the effect of this replacement on the hydrogen production cost was also evaluated. In this case, a sensitivity analysis was performed considering that catalyst replacement will vary depending on the operation time. Fig. 10 shows the H_2 production costs due to the replacement of the catalyst at each 5, 10, 15 and 20 years of plant operation. Therefore, the catalyst will be replaced 4, 2, 1 and 1 times, respectively. The results demonstrated that the H_2 production costs decreased from 9.05 to 8.92 US\$/kg, when the period of catalyst replacement increased from 5 to 20 years of plant operation.

Analysis of annual profitability

The minimum hydrogen selling price was obtained considering an NPV of zero (US\$ 10.04/kg H_2). The economic analysis was carried out in order to evaluate the feasibility of a distributed hydrogen production plant considering an average hydrogen market price of US\$ 13.44/kg H_2 . This average hydrogen price was obtained using the minimum and maximum H_2 market selling prices from an annual report for hydrogen refueling stations in California for the third quarter of 2017 [44]. This report is annually renewed by State of California Energy Commission and California Air Resources Board in order to establish a network of publicly available hydrogen refueling stations to support the fuel cell electric vehicle (FCEV) market.

For the analysis of annual profitability, the following assumptions were taken into account: a financing plan with an equity cost of 10%, a debit cost of 1.05% and funding of approximated US\$ 11.9 million (80% of the total amount of total capital cost). Fig. 11 show the NPV versus interest rate, considering a financing period of 10 y for a hydrogen selling price of US\$ 13.44/kg H_2 . The results revealed that the processes exhibited an NPV around US\$ 18.0 million, with an equity cost of 10%. The value of the IRR was 42.40%. These results highlight the economic feasibility of the process.

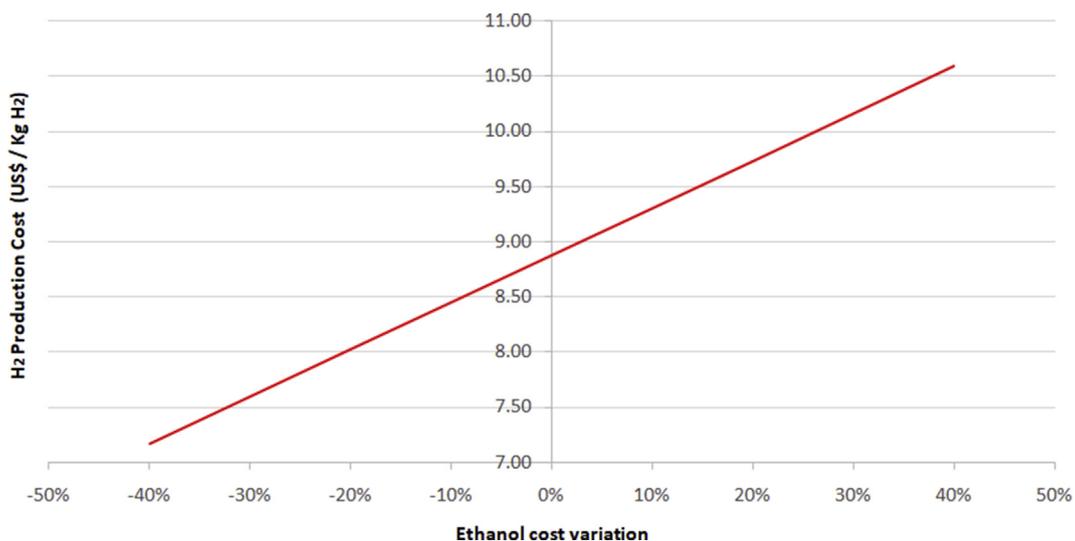


Fig. 8 – Influence of ethanol price on the H_2 cost production.

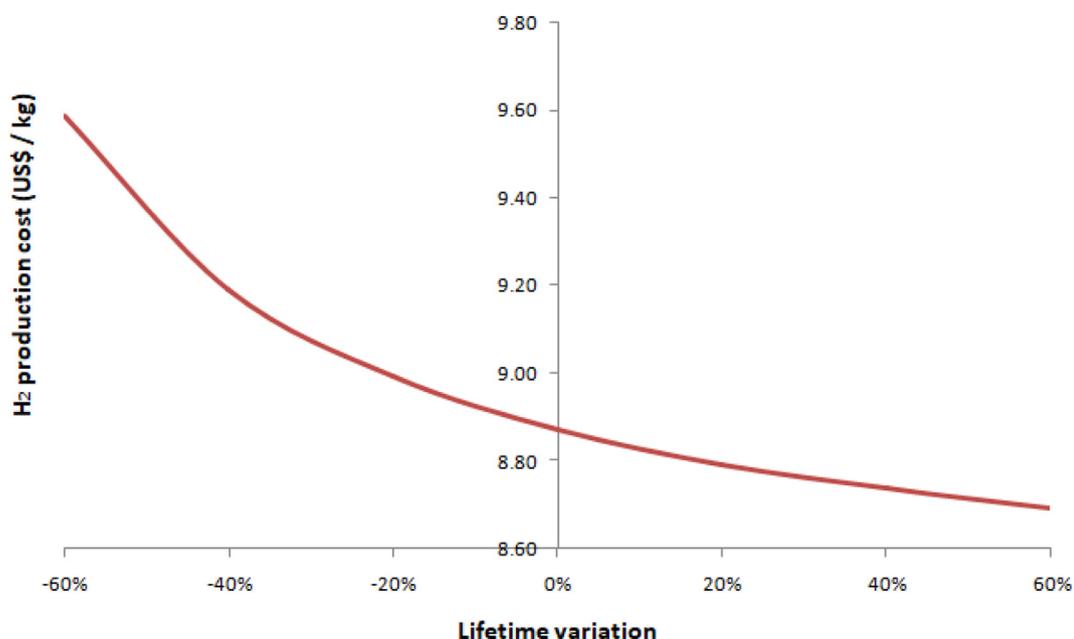


Fig. 9 – Influence of the plant operation lifetime on the hydrogen production cost.

A sensitivity analysis of selling price of hydrogen for the respective IRR and for the payback were calculated (Figs. 12 and 13). From Fig. 12, it is observed that the price of hydrogen in the market (US\$ 13.44/kg H₂) corresponds to an IRR greater than the discount rate, indicating the economic feasibility of the process. In addition, the variation of IRR causes a reduction on the payback (Fig. 13). For the IRR = 42.40%, the payback is 3 years.

Fig. 14 shows the curve of payback as a function of time. The results show that the cost of investment could be pay down after 2 years of operation.

The revenue value as a function of the amount of H₂ produced is presented in Fig. 15. The variable costs are the direct costs and the fixed costs involve the indirect costs and general costs. The plant obtained a positive profit when the H₂ production is higher than 33.3 kg/h (413.97 m³ H₂ produced per 170.17 L ethanol feed).

The demand for H₂ is growing over the years but the growth of existing stations capacity for H₂ supply is not following this demand [44]. For example, in the first quarter of 2017, the stations capacity in California was 4,800 kg H₂/day while the utilization rate was approximately 15%. On the

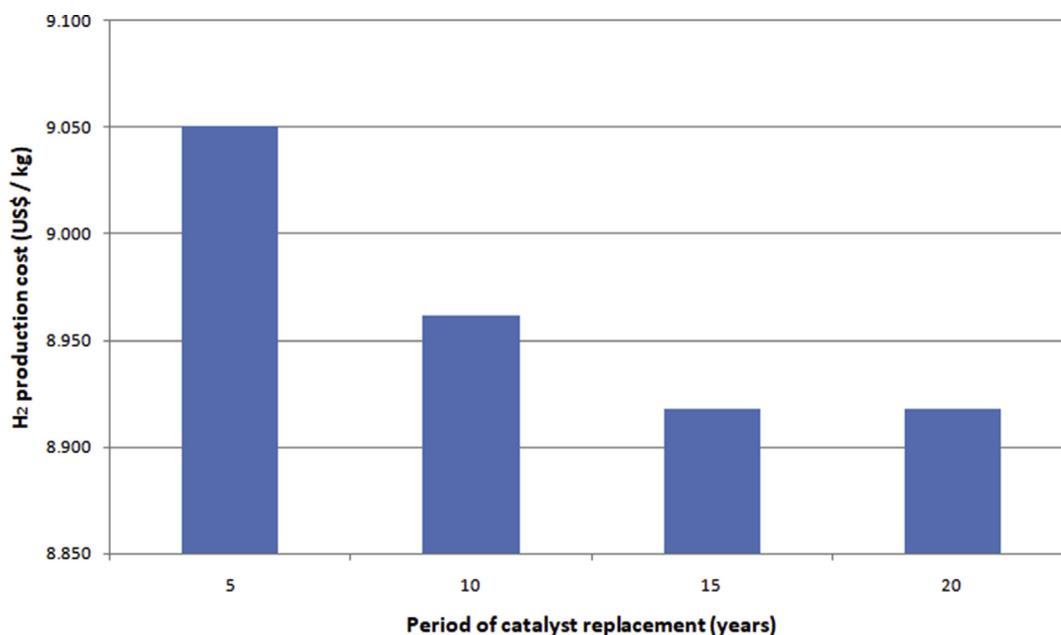


Fig. 10 – Hydrogen production cost as a function of the period of catalyst replacement.

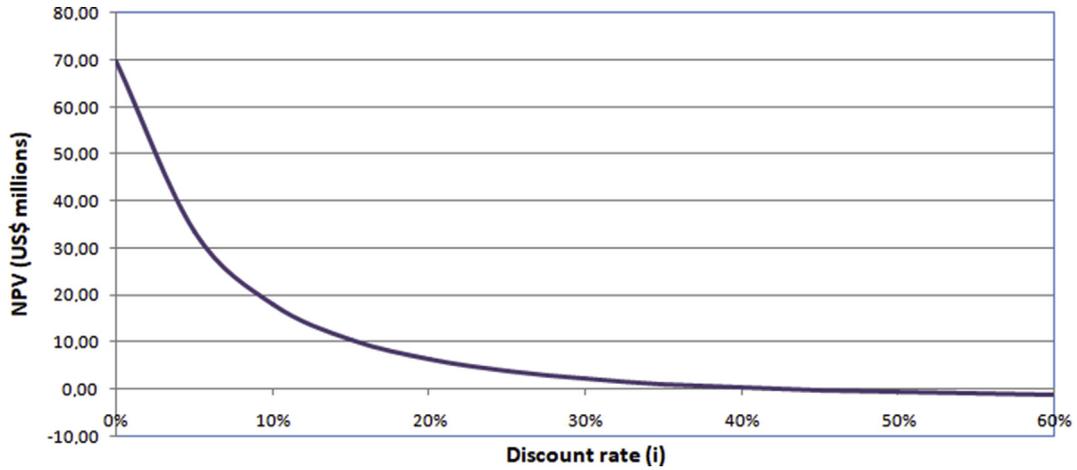


Fig. 11 – Net present value (NPV) versus interest rate.

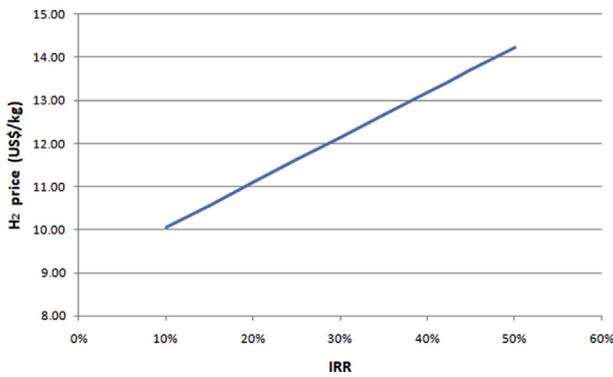


Fig. 12 – Hydrogen selling price for the IRR.

other hand, in the third quarter of 2017 (last published case), the capacity was 5,750 kg H₂/day and the utilization rate was approximately 27%. If this trend continues, the demand for H₂ may exceed the installed capacity, and may result in limiting the growth of the electric cars driven to fuel cells and therefore, their penetration into the market. Therefore, it is necessary to invest in compact and local means of production.

Considering the increasing demand, a utilization rate greater than approximately 25% (referring to breakeven value of 34.0 kg/h with a production capacity of 130.0 kg/h) guarantees a positive NPV for implementation of this technology for local H₂ production in fuel stations. Thus, these results suggest that the price of hydrogen produced from fuel processor based on steam reforming of ethanol for a refueling station using monolith reactors is quite competitive compared

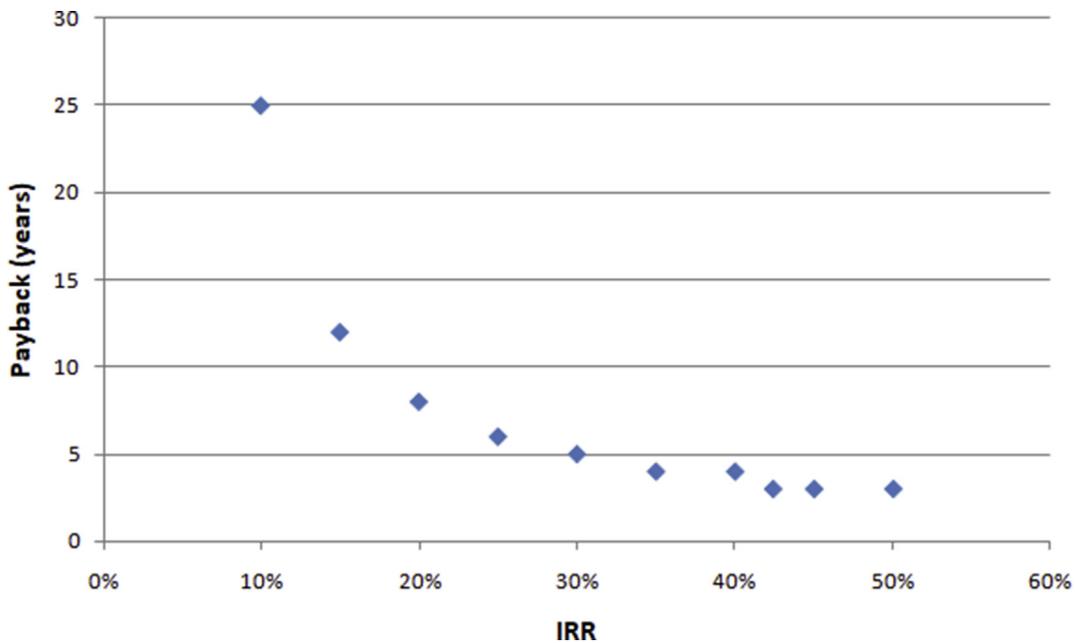


Fig. 13 – Payback period for the IRR related to the selling price of hydrogen.

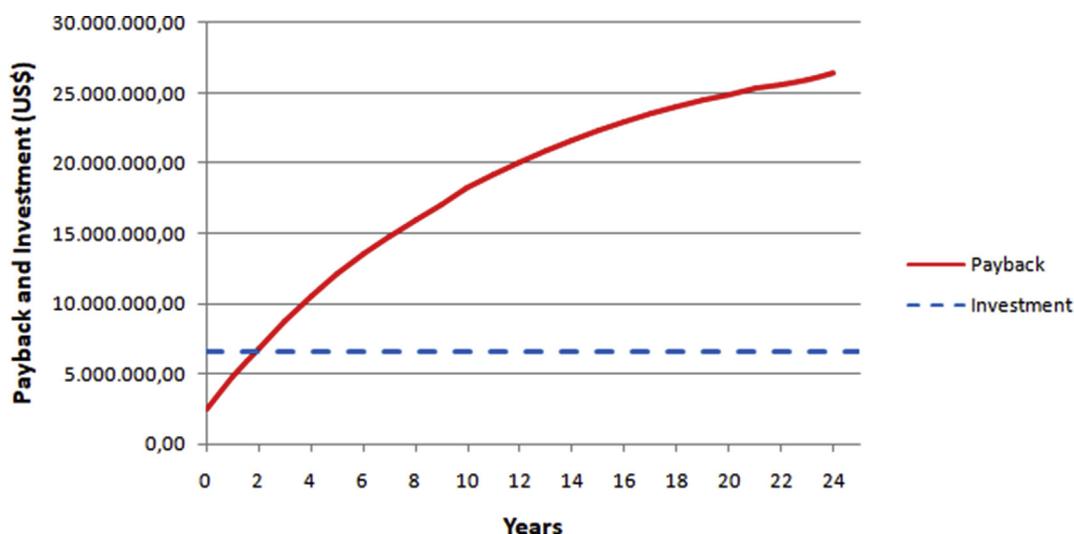


Fig. 14 – Payback over time.

to the hydrogen market price practiced in the hydrogen refueling stations in California.

In the literature, there are several studies about the techno-economic analyses of H_2 production using other renewable sources such as solar and wind [46–49]. Zhao and Brouwer [46] reported an economic analysis for self-sustainable HRS using onsite electrolysis integrated with wind and solar energy sources and considering a hydrogen production of 25 kg H_2 /day. They obtained a H_2 production cost of 6.71 and 9.14 US\$/kg H_2 for the wind and solar powered stations, respectively. Yadav and Banerjee [47] reported an economic assessment of hydrogen production from solar driven high-temperature steam electrolysis process with a plant scale of 0.2 kg H_2 /s (17,280 kg H_2 /day). For a photovoltaic (PV) power plants, they obtained a levelized cost of H_2 production between 16 and 22 US\$/kg H_2 , depending on operating conditions. Shaner et al. [48]

reported a techno-economic analysis of photo-electrochemical (PEC) and photovoltaic-electrolytic (PV-E) solar hydrogen production of 10,000 kg H_2 /day. The levelized costs for the hydrogen production was 11.4 and 12.1 US\$/kg H_2 for the PEC and PV-E systems, respectively. In the case of wind electrolysis, for a plant size scalable from 1,000 kg/day to the 50,000 kg/day and considering wind classes 3–6 across the U.S., the costs of hydrogen production ranged from 3.74 to 5.86 US\$/kg H_2 [49]. A comparison between these results with that obtained in this work showed that the levelized costs of hydrogen production from bioethanol (US\$ 10.04/kg H_2) was close to that observed for solar electrolysis, but it is still higher than that obtained for wind electrolysis. However, for all renewable sources, the costs are not competitive with that one for hydrogen production from steam reforming of natural gas (1.39 US\$/kg H_2) [48].

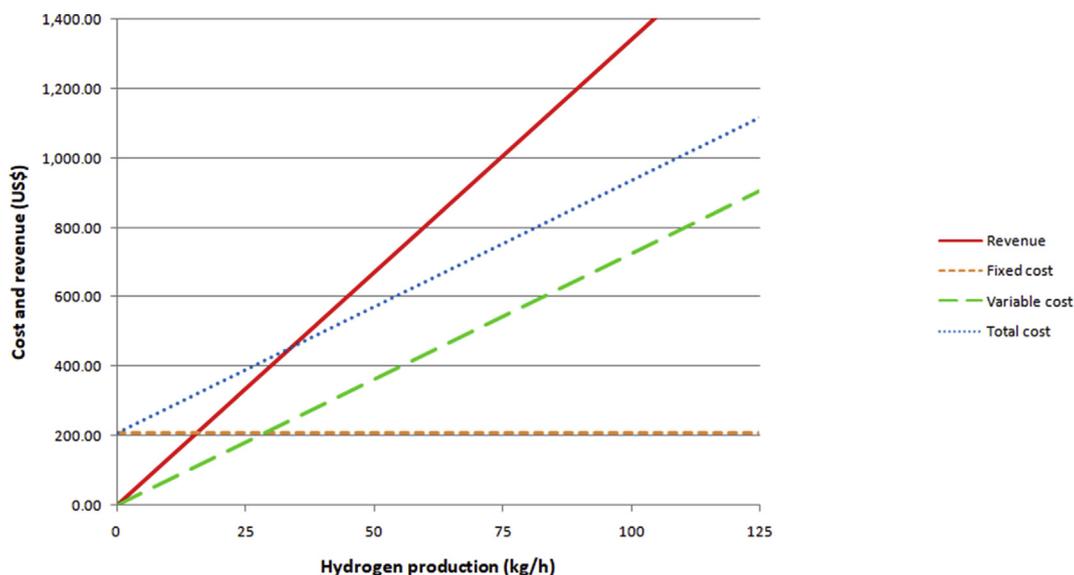


Fig. 15 – Breakeven chart of H_2 production.

Conclusions

The hydrogen selling price for H₂-powered car refueling stations calculated from our experimental data obtained using monolithic catalysts to simulate a compact fuel processor was evaluated. The contribution of different components costs to the hydrogen production cost was determined. A sensitivity analyses was also performed examining the effect of ethanol cost.

Costs with catalysts accounted for only 0.5% of the production hydrogen cost, even using noble metal catalysts. In addition, the reforming section costs using monolith reactors should be lower than those obtained with conventional reactors, since catalysts represents around 20% of the reforming section costs. On the other hand, the ethanol cost contributed to 46.1% of the production hydrogen cost. Even using the expensive Brazilian ethanol price (0.81 US\$/L ethanol), the hydrogen production cost obtained in this work was US\$ 8.87/kg H₂, which is lower than the current market prices (US\$ 13.44/kg H₂) practiced at hydrogen refueling stations in California.

The economic analysis indicates a NPV of US\$ 18.0 million, an IRR of 42.40% and a payback after 2 years using the hydrogen selling price of 13.44 US\$/kg H₂. This result showed that the development of a compact fuel processors using monolith catalysts can be economically feasible to provide hydrogen to H₂-powered cars at competitive costs in refueling stations.

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