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Thermal Efficiency of On-site, Small-scale Hydrogen Production Technologies using Liquid Hydrocarbon Fuels in Comparison to Electrolysis: a Case Study in Norway

Lucrezia Ravasio^a, Silvia Ravelli^{a*}, Mohamad Mustafa^b

^aUniversity of Bergamo, Department of Engineering and Applied Sciences, Marconi St. 5, Dalmine 24044, Italy

^bUiT The Arctic University of Norway, Department of Building, Energy and Material Technology, Lodve Langes Gate 2, Narvik 8515, Norway

Abstract

The main goal of this study was to assess the energy efficiency of a small-scale, on-site hydrogen production and dispensing plant for transport applications. The selected location was the city of Narvik, in northern Norway, where the hydrogen demand is expected to be 100 kg/day. The investigated technologies for on-site hydrogen generation, starting from common liquid fossil fuels, such as heavy naphtha and diesel, were based on steam reforming and partial oxidation. Water electrolysis derived by renewable energy was also included in the comparison. The overall thermal efficiency of the hydrogen station was computed including compression and miscellaneous power consumption.

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Keywords: Hydrogen ; Liquid hydrocarbons; Steam reforming; Partial oxidation; Electrolysis

1. Introduction

The European Union (EU) should reduce dependence on imported oil and cut carbon emissions in transport by 60% by 2050, as established in the White Paper 2015, to keep global warming to within 2°C of pre-industrial times.

* Corresponding author. Tel.: +39 -035-2052346; fax: +39-035-2052077

E-mail address: silvia.ravelli@unibg.it

The more ambitious COP21 commitment to limit temperature rises to 1.5°C would require a complete decarbonization of transport by 2050. The conceived EU strategy includes speeding up the deployment of advanced biofuels, hydrogen and electricity, and accelerating the transition towards zero-emissions vehicles [1]. In particular, fuel cell electric vehicles might have great potential in the long term 2050 contest, provided that hydrogen production and refueling infrastructure is available. Among the Nordic countries, Norway has the potential to become a pioneer in the development of hydrogen supply and infrastructure technologies. In fact, the Norwegian Environment Agency has established the strict 40% emission reduction, compared with the 1990 level, by 2030 [2]. Such an ambitious program is rooted in the access to a wide range of renewable energy sources, which could be exploited for hydrogen production by water electrolysis [3]. In a more general perspective, decentralized production technologies, as an option to exclusively centralized power production, are crucial in Norway due to low population density and the lack of natural gas distribution grid [4]. An alternative to electrolysis is on site hydrogen production by processing liquid fossil fuels currently available at existing filling stations. Indeed, the advantage of existing infrastructures of distribution of gasoline and diesel has promoted active research on liquid hydrocarbon fuel conversion into hydrogen in small scale reactors [5, 6], although liquid fuel reforming is much more challenging than gaseous fuel reforming, because of carbon deposition on catalysts and coke formation [7].

This work attempts to provide a feasibility study for the implementation of on-site hydrogen production from liquid hydrocarbon fuels in Narvik (Norway), through the well-known chemical techniques of steam reforming, partial oxidation and auto-thermal reforming [8]. The target production is 100 kg/day (46.4 Nm³/h) of high purity hydrogen (at least 99.995%), to meet the expected demand in that small city of the Nordland county. The nearest centralized hydrogen production plant is located in Glomfjord, 400 km South of Narvik. Alleviating the need for large scale transport of hydrogen from Glomfjord to Narvik is vital for the hydrogen economy development in Nordland, because of the cold weather and remote location. Actually, recent initiatives and research projects to implement hydrogen in road transport focused on Southern Norway [9]. Water electrolysis has also been included as a benchmark, with the aim of assessing the thermal efficiency of each hydrogen generation process together with the overall efficiency of the small-scale hydrogen station.

2. Small scale, on-site hydrogen station

The main components of a gaseous hydrogen production and dispensing plant for road transport application are presented in Fig. 1. The first block is obviously the on-site hydrogen generator, which should meet the above mentioned requirements in terms of flow rate and purity of the delivered hydrogen stream. Whatever the production technology, the hydrogen is stored in a temporary buffer at relatively low pressure. Thanks to the first compressor unit, hydrogen pressure is increased from the low level of about 10 bar to a medium level of 400 bar. Then hydrogen is stored in a second buffer. From the medium pressure storage, hydrogen can take two paths, either flow through a cascade filling system to a dispensing system for delivery to the users or be sent to the second stage compression, where its pressure is increased from 400 bar up to 750 bar. The high-pressure storage and dispensing unit complete the block diagram. The reason for having medium and high pressure levels is to serve two types of hydrogen fed vehicles: buses and cars, respectively. Practically, a larger number of tanks at medium pressure can be taken on board in a bus; conversely, a smaller number of tanks at high pressure are mandatory in a car, due to the very limited space available. In fact, the pressure of the delivered hydrogen is dictated by the available on-board storage system and the desired mileage of the vehicle between fill-ups.

Hence, leaving aside the hydrogen generator that will be discussed in the next section, three storage units of different size and characteristics are needed to accommodate the unavoidable mismatches between production and demand. The low-pressure storage is a temporary buffer where hydrogen flows before entering the first compressor, whereas medium-pressure and high-pressure storage must be dimensioned according to the demand, since they are connected to the dispensing systems. The major challenge in designing a refueling station is to deal with the highly variable fuel demand. It has been decided to size medium-pressure and high-pressure storage with a nominal capacity of about 30% of the daily demand, as suggested by published literature [10]: actually, 30 kg is enough fuel for a single bus and six cars, approximately. The low-pressure storage should have a capacity of 40 kg. Among the available hydrogen tanks (classified under type I, II, III, IV and V), type I was found to be suitable for the low pressure storage, type III for the medium pressure storage and type IV for the high pressure storage.

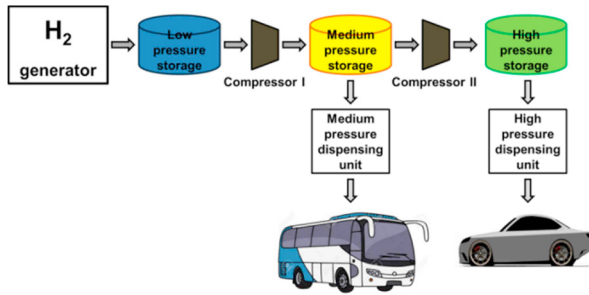


Fig. 1. Main components of a hydrogen production and dispensing plant for road transport applications.

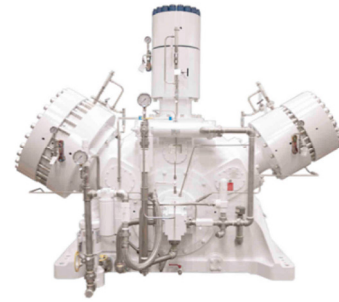


Fig. 2. PDC-SP-900-5000-15000 three-stage compressor for hydrogen gas vehicle filling [11].

A diaphragm compressor, such as that shown in Fig. 2, was chosen for the current application: it is a three-stage compressor manufactured by PDC Machines, Inc., whose discharge pressure can be as high as 950 bar gauge pressure [11]. Conceived to be leak free, each compressor includes a leak detection system.

3. H₂ generation

Four different solutions for 100 kg/day hydrogen production were taken into account drawing inspiration from [12]. Liquid fossil fuels available in a conventional filling station, such as heavy naphtha and diesel, were chosen as inputs. The investigated technologies included steam reforming with and without pre-reforming, auto thermal reforming and partial oxidation. Hydrogen generation from water electrolysis was also considered for comparison purposes.

Fuel chemical characterization, using the PIONA (n-Paraffin, Iso-paraffin, Olefin, Naphthene, and Aromatic) approach and higher heating value (HHV) is listed in Table 1: please note that both fuel streams were assumed to be sulfur-free. The transportation fuel conversion processes have been modeled using the chemical process simulator software VMGSimTM 7.0. All reactors were solved iteratively by Gibbs minimization calculation based on atom balances as constraints. The equilibrium constant was computed from the ideal gas Gibbs free energy for each compound in the reaction. This method offers the advantage of not necessarily knowing the chemical reactions involved. Each Gibbs minimization reactor was modelled as an isothermal process, at the operating temperature specified in Table 2. The following major reactors were simulated:

- Steam reformer (SR): liquid fuel and on-site generated steam react to provide syngas, which is a mixture of carbon monoxide and hydrogen, as shown in



It is a highly endothermic catalytic reaction, so heat is required from an external source. Convergence of the solution was verified against imposed reactor temperature (815.6°C) and pressure drop. The same reaction takes place in the pre-reformer reactor, but at lower temperature (510°C), together with methanation exothermic reaction.

- Shift reactor: carbon monoxide is converted into carbon dioxide through the following exothermic reaction



The operating temperature is 350°C in case of High Temperature (HT) shift reactor. CO concentration is expected to fall from 10-15 mol%-dry to 1-2 mol%-dry. The same reaction is replicated at 204.4°C in the Low Temperature (LT) Shift Reactor, with a lower conversion rate. This step should guarantee a reduction in CO concentration from 1-2 mol% to 0.1-0.2 mol%.

- Auto-thermal reformer (ATR): the liquid fuel undergoes both a partial oxidation thanks to the oxygen contained in the inlet air stream and a reforming reaction with the water steam. The final product is again a mixture of carbon monoxide and hydrogen. The reactor temperature is that of the steam reformer (815.6°C). Heat for the endothermic reaction is provided by internal heat recovery.
- Partial oxidation (POX) reactor

Table 1. Liquid hydrocarbon fuel specification.

<i>PIONA</i>	Heavy Naphtha	Diesel
Aromatics (vol%)	8,3	20
Naphthenes (vol%)	38	25
i-Paraffins (vol%)	41.5	22
Paraffins (vol%)	8	32
Olefins (vol%)	3	-
H/C ratio	1.96	1.89
Higher Heating Value (kJ/kg)	45604	46199
Density @15°C (kg/m ³)	754.3	830.6

Table 2. Operating temperature of major reactors.

Reactor	Temperature (°C)
Steam reformer	815.6
Pre Reformer	510.0
HT Shift	350.0
Auto-thermal reformer	815.6
LT shift	204.4
Partial Oxidation Reactor	1200.0
Pressure swing absorption H2 side	70.0

The following exothermic reaction between liquid fuel and pure oxygen occurs:



The operating temperature, equal to 1200°C, is higher than that of the steam reformer to compensate for the lack of catalyst.

Gas composition at reactor outlet is at chemical equilibrium: the assumption is well-founded if the reactor is operated at high enough temperature, thus ensuring high rate of reaction [6]. Whatever the production chain, hydrogen is separated from carbon dioxide and other inert gases by means of two techniques: pressure swing absorption (PSA), in which the syngas flows through an absorber bed that entraps unwanted gaseous components, or polymeric membrane separation, in which selective diffusion of hydrogen takes place. Concerning the PSA, the operating pressure is between 1.35 and 10 bar. The PSA off-gas is used for the combustor, thus providing the heat required by the reformer. The combustor is operated in an adiabatic mode. If the heat demand of the steam reforming cannot be met by burning the off-gas, additional fuel is fed to the burner.

The steam reforming process is shown in Fig. 3a. The liquid fuel feed is taken from storage (FuelFeedStorage); the pump P1 increases its pressure before entering a SR reactor to obtain syngas. The syngas stream flows through a cooler (C1) before entering the HT shift reactor. Then it passes through a second cooler (C2) to accomplish a proper inlet temperature for the PSA unit. The off-gas from the PSA is burned in a combustor, with pressurized ambient air. Waste gases flow into two coolers (H1, C3), before being released into the environment, in order to provide heat for reforming and steam generation, respectively. In fact, the required steam flow rate is generated on-site from feed water at ambient conditions: heat is provided not only by C3 but also by C1. The component named “CO1” is a Claus Oxygen Calculator, useful for setting the oxygen fraction excess in the combustor so as to obtain the desired inlet air flow rate.

Pre-reforming followed by steam reforming is depicted in Fig. 3b. The liquid fuel, after compression in the pump P1, is sent to the pre-reformer where reacts with steam to provide a methane-rich gas, which flows into the reformer reactor. First cooler C1, HT shift reactor and second cooler C2 are placed downstream of the reformer. Finally, hydrogen is separated from other gases in the PSA unit, similarly to what is shown in Fig. 3a. The off-gas mixes with the pressurized ambient air in M2 before entering the combustor. CH1 and CH2 extract heat from the waste gases to supply heat to the reformer and pre-reformer, respectively. Heat needed to convert feedwater to steam is provided by cooler C3. Again, CO1 is used to regulate the oxygen content in the combustor.

Fig. 3c shows the process flow diagram of the auto thermal reforming. The compressed liquid fuel is reformed with steam in the Auto Thermal Reformer. The produced syngas passes through a first cooler (C1), the HT shift reactor, a second cooler (C2) and the low temperature (LT) shift reactor to generate hydrogen-rich gas. This enters a third cooler (C3) before being separated into pure hydrogen and off-gas in the PSA unit. Since the LT shift promotes CO conversion into H₂, the off-gas flow rate exiting the PSA is minimized. The off-gas is mixed (M3) with pressurized ambient air and burned in a combustor: heat generated is primarily used to vaporize the liquid fuel for the ATR (heater H1) and, secondarily, to convert feedwater into steam (cooler C4).

Partial oxidation is illustrated in Fig. 3d: the liquid fuel is sent to a very high temperature reactor for partially oxidization with oxygen, without using a catalyst. The outlet syngas is cooled in C1 before entering the HT shift reactor, a second cooler C2 and the LT shift reactor, to minimize the off-gas production. Finally, the syngas passes through a membrane separator to obtain high purity hydrogen. The high temperature off-gas is sent to a combustor where it reacts with oxygen, instead of ambient air.

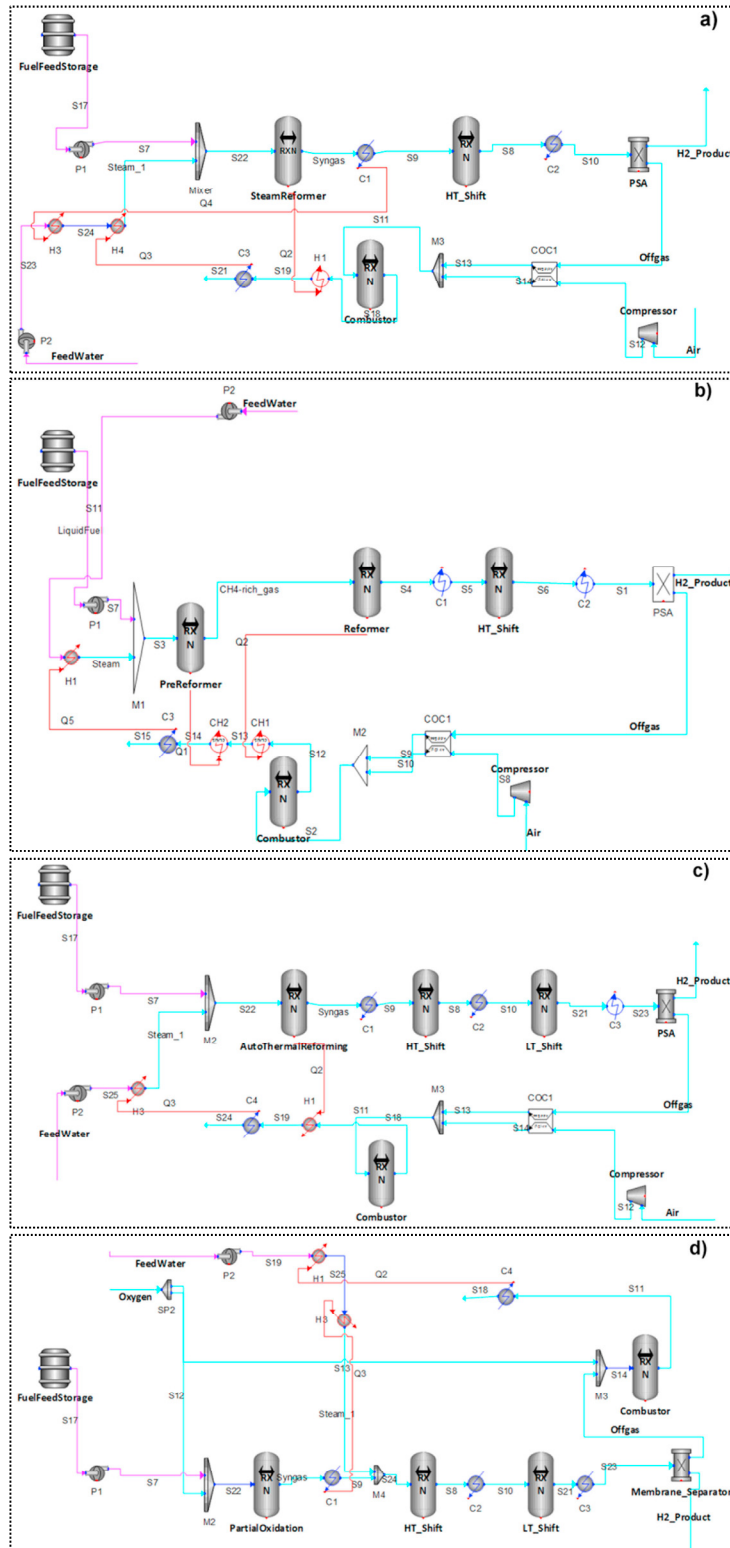


Fig. 3. Process flow diagram of the investigated solutions for H₂ generation from liquid hydrocarbon fuels: a) steam reforming; b) pre-reforming followed by steam reforming; c) auto thermal reforming; d) partial oxidation.

In fact, the novelty in this layout is the oxygen stream: one fraction (S12) feeds the PO reactor, the other one (S13) the combustor. A cooler (C4) is located downstream of the combustor for heat recovery from flue gas (S11). This cooler (C4), together with that located downstream of the POX reactor (C1), provides the heat for steam generation. Supplemental fuel firing could help to generate steam.

As far as water electrolysis is concerned, an electrolyser receives water and electricity as inputs and gives back gaseous hydrogen and oxygen as outputs. Water electrolysis works well at small scales and can be considered more sustainable by using renewable electricity. For the current application, electricity would be provided by Narvik Energi, a power company that serves Narvik: it operates seven hydroelectric power stations and a wind farm in Nordland County. The feedwater purification system is also required and typically included in the electrolyser unit. The HySTAT®-60-10 alkaline water electrolyser, manufactured by Hydrogenics, is fit for the purpose: it can produce 100 kg/day of high purity hydrogen (up to 99.998%) at 77% of its nominal capacity, with an estimated power consumption of 5785 kWh/day and tap water usage of 1890 l/day.

3.1. Thermal efficiency of the H₂ generation chain

The performance summary of the investigated five methods for on-site hydrogen generation is presented in Table 3. The thermal efficiency η_{th} of each production chain has been calculated as the ratio between output power and input power, as follows

$$\eta_{th} = \frac{m_{H_2} HHV_{H_2}}{m_{fuel} HHV_{fuel} + \frac{P_{el}}{\eta_{el}}} \quad (4)$$

The amount of hydrogen produced is $m_{H_2} = 100$ kg/day, with HHV_{H_2} of 142 MJ/kg, starting from the fossil fuel flow rate m_{fuel} , with HHV_{fuel} indicated in Table 1. P_{el} refers to the electrical power consumption of pumps, air compressors or electrolyser. It was assumed that the required electric power is imported from hydropower stations at efficiency of $\eta_{el} = 0.90$. In fact, the efficiency of the Norwegian power sector is, on average, close to 97% since hydropower is the dominant mode of electricity generation; transmission and distribution loss percentage is approximately 7% of total electricity disposition [13]. Eq. 4 provides a simple but an effective term of comparison for finding the highest performing system on an equal basis, i.e. the same dispensing capacity. The efficiency ranking among the four options based on hydrocarbons is not affected by the fuel considered. However, processes using heavy naphtha as input resulted in a slightly higher efficiency than those using diesel. This is consistent with the fact that heavy naphtha has slightly higher H/C ratio than diesel (the higher the H/C ratio the lower the carbon dioxide formation). Steam reforming from heavy naphtha was found to be the most efficient solution with $\eta_{th} = 78\%$, followed by partial oxidation ($\eta_{th} = 67\%$). It is not worthwhile to include pre-reforming since η_{th} decreases at 59%. Electrolytic production of hydrogen is performed with η_{th} of 61% and 0.079 m³/h water consumption.

Table 3. Performance summary of hydrogen production chains with target capacity of 100 kg/day.

	Steam reforming	Pre-reforming & steam reforming	Auto thermal reforming	Partial oxidation	Electrolysis
m_{H_2} (kg/day)			100 kg/day		
$m_{H_2} HHV_{H_2}$ (GJ/h)			0.59		
<i>Heavy Naphtha</i>					
m_{fuel} (kg/day)	393.6	520.8	537.6	468.0	-
$m_{fuel} HHV_{fuel}$ (GJ/h)	0.75	0.99	1.02	0.89	-
Water consumption (m ³ /h)	0.058	0.061	0.083	-	0.079
Power consumption (kW)	2.56	4.46	4.61	0.14	241.06
Thermal efficiency (%)	78.0%	58.7%	56.9%	66.5%	61.4%
<i>Diesel</i>					
m_{fuel} (kg/day)	398.4	523.2	542.4	465.6	-
$m_{fuel} HHV_{fuel}$ (GJ/h)	0.77	1.01	1.04	0.90	-
Water consumption (m ³ /h)	0.059	0.063	0.089	-	0.079
Power consumption (kW)	2.63	4.51	4.78	0.14	241.06
Thermal efficiency (%)	76.1%	57.7%	55.6%	66.0%	61.4%

It is important to notice that the reported η_{th} values are inevitably affected by η_{el} in eq. 4: η_{th} increases with η_{el} , as depicted in Fig. 4. This is particularly evident for the most energy-consuming process, such as electrolysis, whose thermal efficiency can reduce from $\eta_{th} = 68\%$ at $\eta_{el} = 1$ to $\eta_{th} = 24\%$ at $\eta_{el} = 0.35$. In the former case, η_{th} would coincide with the common definition of HySTAT[®]-60-10 electrolyser efficiency; in the latter case η_{th} would be vitiated by the use of electrical power generated by fossil fuel power stations, whose average efficiency ranges from 0.35 to 0.45 [14]. Figure 4 suggests that the sustainability of water electrolysis, as compared to competitors, does not allow $\eta_{el} < 0.8$. Results obtained using heavy naphtha as feedstock were compared against those from [12] for 1000 kg/day hydrogen generation, in order to assess scale effects on thermal efficiency (Fig. 5). Please, note that data contained in [12] have been recalculated to account for the current assumption of η_{el} equal to 0.90, instead of 0.38. Steam reforming showed almost constant thermal efficiency, whatever the capacity, thus confirming excellent down-scaling capability. The same is valid for the autothermal reforming which combines partial oxidation and steam reforming. Conversely, partial oxidation alone was affected by 5%-points loss in thermal efficiency when reducing the capacity by an order of magnitude.

4. Overall thermal efficiency of the H₂ station

The overall performance of the hydrogen station is summarized in Table 4. The components located downstream of the hydrogen generator (see Fig. 1) have been accounted for to get the overall thermal efficiency. Accordingly, the term P_{el} of eq. 4 includes the following:

- electric power consumption for hydrogen compression, assuming an isentropic efficiency of 65%, given the small designed flow rate [15];
- miscellaneous power consumptions, assumed to be 5% of those of a plant producing 1000 kg/day of hydrogen;
- power requirement for on-site oxygen generation, in case of partial oxidation, which is equal to 460 kWh/ton of oxygen delivered, based on [12].

The highest overall efficiency of 59% or 58% can be achieved by steam reforming of heavy naphtha and diesel, respectively. Partial oxidation is also a viable solution ensuring a global efficiency of about 51%. In case of water electrolysis, the overall performance is 49%. With reference to hydrogen production from heavy naphtha, once again results were compared with those from [12] for 1000 kg/day hydrogen station (Fig. 6). It should be noted that the hydrogen station conceived here has higher storage pressure levels than those assumed in [12]: 400 bar instead of 173 bar at the medium pressure storage, and 750 bar instead of 432 bar at the high pressure storage. The heavier burden of hydrogen compression, due to higher pressure ratio, is the main reason behind the penalty in the overall thermal efficiency associated with the smaller capacity of 100 kg/day, whatever the production chain. Moreover, it is presumed that the current compression efficiency of 65% is lower than that used in [12], which is unfortunately unreported. For the most promising process, i.e. steam reforming from heavy naphtha, the overall performance falls from 71% to 59%. The overall thermal efficiency drop is roughly the same (-13% points) for partial oxidation.

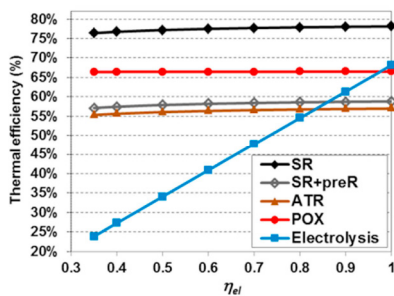


Fig. 4. Hydrogen generation thermal efficiency as a function of power station efficiency η_{el} (fuel: heavy naphtha).

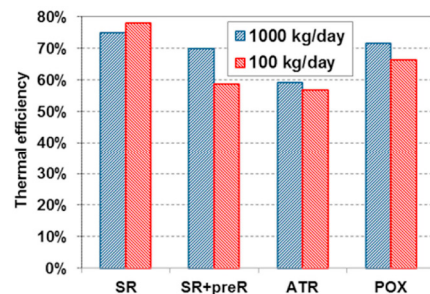


Fig. 5. Hydrogen generation thermal efficiency for capacity of 100 kg/day vs. 1000 kg/day [12] (fuel: heavy naphtha).

Table 4. Performance summary of hydrogen stations with target capacity of 100 kg/day.

	Steam reforming	Pre-reforming & steam reforming	Auto thermal reforming	Partial oxidation	Electrolysis
m_{H_2} (kg/day)			100 kg/day		
$m_{H_2} HHV_{H_2}$ (GJ/h)			0.59		
Heavy Naphtha					
$m_{fuel} HHV_{fuel}$ (GJ/h)	0.75	0.99	1.02	0.89	-
Water consumption (m ³ /h)	0.058	0.061	0.083	-	0.079
Total import P_{el} (kW)	62.81	64.71	64.86	66.69	301.31
H ₂ generation	2.56	4.46	4.61	0.14	241.06
H ₂ compression	60.0	60.0	60.0	60.0	60.0
O ₂ generation	-	-	-	6.3	-
Miscellaneous	0.25	0.25	0.25	0.25	0.25
Overall thermal efficiency (%)	59.2%	47.4%	46.2%	51.2%	49.1%
Diesel					
$m_{fuel} HHV_{fuel}$ (GJ/h)	0.77	1.01	1.04	0.90	-
Water consumption (m ³ /h)	0.059	0.063	0.089	-	0.079
Total import P_{el} (kW)	62.88	64.76	65.03	66.69	301.31
H ₂ generation	2.63	4.51	4.78	0.14	241.06
H ₂ compression	60.0	60.0	60.0	60.0	60.0
O ₂ generation	-	-	-	6.3	-
Miscellaneous	0.25	0.25	0.25	0.25	0.25
Overall thermal efficiency	58.1%	46.7%	45.4%	50.9%	49.1%

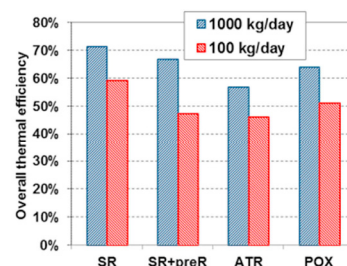


Fig. 6. Hydrogen station overall thermal efficiency for capacity of 100 kg/day vs. 1000 kg/day [12] (fuel: heavy naphtha).

5. Conclusions

The design of a small-scale, on-site hydrogen production and dispensing plant for road transport applications was conceived, for a target capacity of 100 kg/day. The selected context for this study is the city of Narvik, Norway. Four different processes to generate hydrogen from liquid hydrocarbon fuels (heavy naphtha and diesel) were compared with water electrolysis, in terms of thermal efficiency. Moreover, the overall thermal efficiency of the hydrogen station, which consisted of hydrogen generator, compressors and storage tanks at three pressure levels (10, 400 and 750 bar), was computed to find the most promising solution, under the assumption that required electricity is generated from hydropower stations available in Norway. Steam reforming of heavy naphtha ensured the highest overall efficiency of $\eta_{th} = 59\%$, followed by partial oxidation ($\eta_{th} = 51\%$) and water electrolysis ($\eta_{th} = 49\%$).

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