

## POTENTIAL ROLE OF EXERGY IN ANALYSIS OF HYDROGEN INFRASTRUCTURE

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### Abstract

The objective of this paper is to demonstrate the potential role of exergy (second-law) analysis, as a complementary tool for economic assessments of hydrogen infrastructures. Comprehensive thermodynamic analysis is particularly valuable for evaluating hydrogen production technologies that are energy intensive and represent a key infrastructural component.

This study focuses on three selected distributed hydrogen production technologies: natural gas steam reforming, ethanol steam reforming, and electrolysis. The prospect of distributed systems for early market adoption in the absence of a well-developed hydrogen distribution network is a compelling reason for this selection. The thermodynamic performance of these systems are evaluated and discussed in conjunction with their energy costs as major contributors to the total hydrogen production costs. Analyses are also performed in the context of the source energy and feedstock.

For these systems operating under prescribed conditions, the differences between the energetic and exergetic efficiencies are found to be rather insignificant. In light of the calculated unused/destroyed exergies, the limitations for potential hydrogen cost reduction via energy savings are discussed as well.

### 1. Introduction

The potential benefits of hydrogen as an alternative fuel and the prospects of a growing demand advocate development of viable hydrogen infrastructures [1 and 2]. As with any energy carriers, the key components for a hydrogen infrastructure are production, storage, and distribution/delivery, all of which pose technical and economic challenges to successful deployments. The production aspect is perhaps the most important of all as it encompasses issues, such as availability of source energy (feedstocks), fuel utilization efficiency, and environmental effects, among others. However, the ultimate contribution of this alternative fuel to the future energy market will depend on the overall performance of the nascent infrastructure, not just its constituents.

Small-scale distributed hydrogen production technologies can be of particular interest to the industry and other stakeholders because of (1) their minimal dependency (if any at all) on the existence of distribution facilities and (2) an early investor perception of a more manageable risk associated with the technological maturity and the uncertainties

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with the future energy market. Drawing from these attributes, the analytical study presented in this paper focuses on three distributed (forecourt) production technologies without carbon sequestration: natural gas steam reforming, ethanol steam reforming, and electrolysis. The rationale for this selection is as follows:

- The natural gas (methane) reforming technology is matured and a good candidate for early market adoption given the well-developed natural gas infrastructure.
- For the case of ethanol reforming, the feedstock can be produced via a mix of fossil and renewable fuels, mitigating the demand for the fossil fuel. Furthermore, as a liquid, ethanol can be transported to hydrogen production sites without some of the drawbacks inherent in transporting gaseous fuels, especially hydrogen.
- By producing hydrogen as a storage medium, electrolysis can help overcome the intermittency effects of wind and photovoltaic (PV) power generation by closing the temporal gap between the supply and demand for energy.

The purpose of this paper is to explore the potential role of an exergetic analysis in economic assessment of hydrogen production systems that constitute a key infrastructural element. To accomplish this, the cost data were obtained from the available case studies facilitated by the U.S. Department of Energy (DOE) H2A Production Analysis tool [3] for the three aforementioned distributed (forecourt) technologies. H2A Production is a model developed for cost analysis of forecourt and central hydrogen production systems. The forecourt systems, which are the focus of this study, have a capacity of 100 to 1,500 kg of hydrogen per day. The hydrogen produced at an elevated pressure is further compressed for storage and dispensing. Detailed information on the methodology, assumptions, and case studies of H2A are available in the cited reference [3].

Figures 1 and 2, respectively, illustrate the simplified schematics of the steam reformers and the electrolyzer analyzed in this study. In the interest of consistency, efforts were made to emulate the key features of the respective H2A systems, including the operating conditions, in the thermodynamic assessments. The hydrogen leaving the depicted systems corresponds to the product stream of the H2A model upstream of the dispensing/delivery components (compressors and storage units), which are not addressed in this paper. Both energy (first-law) and exergy (second-law) efficiencies of the systems were evaluated and discussed for the designated control volumes (Figures 1 and 2). The energy efficiencies were evaluated based on (1) the delivered electrical energy and pure feed and (2) the source energy for the electricity and for processing the raw feedstock. The latter did not apply to natural gas.

For the thermodynamic analyses, the following assumptions were made:

- An overall efficiency of approximately 32% for electrical power generation and transmission.

- Reference environment temperature ( $T_o$ ) and pressure ( $P_o$ ) of 298 K (25°C) and 1 atm (101.325 kPa).
- Treatment of natural gas, hydrogen, and carbon dioxide as ideal gases.
- A net energy ratio of 1.5 for production of ethanol (i.e., producing 1.5 kJ ethanol per kJ of fossil fuel) – based on a literature review [4, 5, and 6].
- The reactants enter the system at the reference environment conditions ( $T_o, P_o$ ), and the products leave at temperature  $T_o$  and pressure  $P$  ( $P > P_o$ ), which is taken as the operating pressure in the H2A model.

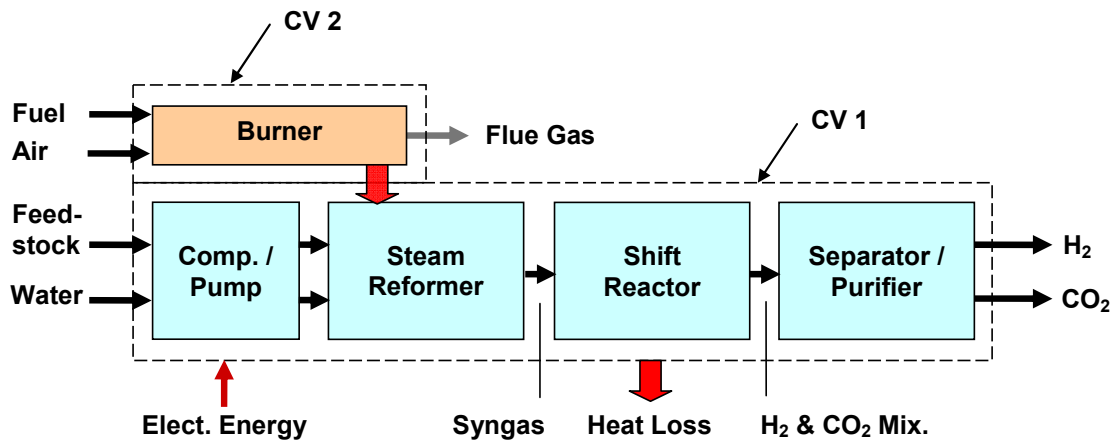


Figure 1. Schematic of natural gas and ethanol reformers.

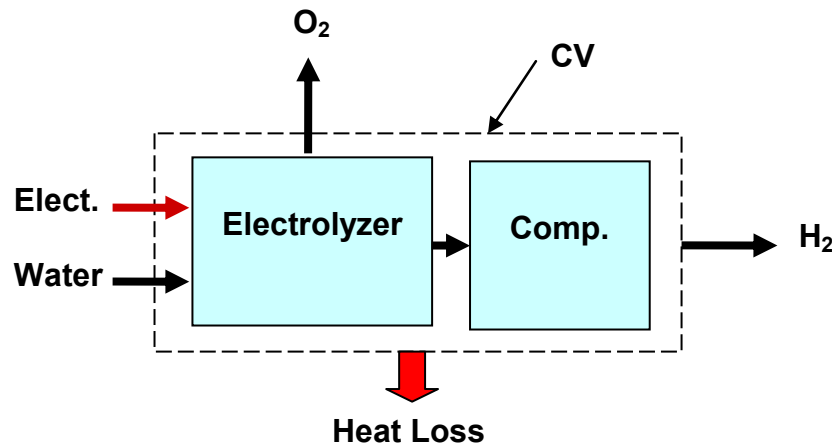
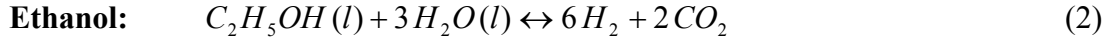
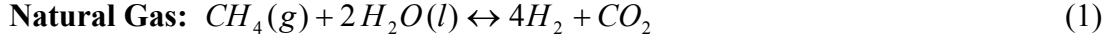


Figure 2. Schematic of electrolyzer.

## 2. Chemical Reactions

The overall chemical reactions for natural gas and ethanol steam reforming are as follows:



The combined reaction for each reformer consists of a strongly endothermic reforming process and a moderately exothermic water–gas shift reaction. The endothermic reaction takes place at a high temperature and pressure. The operating conditions used in the H2A case studies for reforming (850°C and 20 atm) are within the typical ranges [1, 7, and 8]. However, appropriate operating conditions for the reformer and shift reactor of a production plant are determined in part based on the catalyst type and volume. For example, studies have shown that relatively high yields of hydrogen can be obtained at significantly lower temperatures using special catalysts for ethanol reforming [9, 10, and 8].

For reaction (1), a minimum heating amount of 253 MJ per kmol of methane (enthalpy of reaction,  $\Delta H_r^\circ$ ) is required at the standard conditions:  $T_o$  (298 K) and pressure  $P_o$  (1atm). For the ethanol reforming, reaction (2), 348 MJ per kmol of ethanol is required. To meet these heating requirements, the necessary combustion fuel for the burner (CV 2 in Figure 1) is 0.32 kmol/kmol of the feed for the natural gas reformer and 0.28 for the ethanol. These estimates are based on the lower heating value (LHV) of the combustion fuel and the assumption that the fuel is the same as the feed (i.e., natural gas or ethanol). (The theoretical combustion fuel requirement for the natural gas reformer is in good agreement with the estimate of 33% reported by Rosen [9]). The total hydrogen production rates derived from the H2A cases for the natural gas and ethanol reformers are, respectively, 2.43 and 3.49 kmol per kmol of total fuel (feed and combustion fuel combined). The hydrogen yield per kmol of the feed is about 3.2 for the natural gas reformer and 4.47 for the ethanol. These estimates were derived from the reported H2A input and output energy fluxes in conjunction with the respective lower heating values.

For the electrolyzer, the chemical reaction is



## 3. Exergy Analysis

Applying the second law of thermodynamics to the hydrogen production system (CV 1) in Figure 1 yields the following steady-state exergy balance:

$$\int (1 - T_o / T_b) \delta \bar{q} + \bar{x}_{elec.} + \bar{x}_{sep} + \bar{x}_F + a\bar{x}_{H_2O} - b\bar{x}_{H_2} - c\bar{x}_{CO_2} = \bar{x}_{cons.} \geq 0 \quad (4)$$

In this equation,  $\bar{x}_F, \bar{x}_{H_2O}, \bar{x}_{H_2}$ , and  $\bar{x}_{CO_2}$  are the flow exergies of the fluids crossing the system boundary on molar basis, expressed in terms of kJ per kmol of entering fuel (feed). Neglecting the kinetic and potential energy effects, the flow exergy on a molar basis is the sum of chemical exergy,  $\bar{x}^{ch}$ , and thermo-mechanical exergy,  $\bar{x}^{thm}$ , [11]:

$$\bar{x} = \bar{x}^{ch} + \bar{x}^{thm} = \bar{x}^{ch} + (\bar{h} - \bar{h}_o) - T_o(\bar{s} - \bar{s}_o) \quad (5)$$

The coefficients  $a, b, \text{ and } c$  in Equation (4) respectively correspond to the molar quantities of water, hydrogen, and carbon dioxide in the actual chemical reactions. The first term on the left of Equation (4) represents the algebraic sum of exergy flows accompanying the heat transfer crossing the system boundary:

$$\int (1 - T_o / T_b) \delta \bar{q} = \left[ \int (1 - T_o / T_b) \delta \bar{q} \right]_{Endothermic} + \left[ \int (1 - T_o / T_b) \delta \bar{q} \right]_{System \text{ Cooling}} \quad (6)$$

In this equation, the second term on the right has a negative value and denotes the unused thermal exergy that leaves the system via active and passive cooling. The minimum exergy required for separating  $CO_2$  from the  $CO_2/H_2$  mixture is approximated by the following expression [12]:

$$\bar{x}_{sep.} = \bar{R}T_o \left[ \frac{n_{H_2}}{n_m} \ln \frac{n_m}{n_{H_2}} + \frac{n_{CO_2}}{n_m} \ln \frac{n_m}{n_{CO_2}} \right] \quad (7)$$

The parameters  $n_{H_2}$ ,  $n_{CO_2}$ , and  $n_m$  denote, respectively, the number of moles of hydrogen, carbon dioxide, and the mixture in the separator, neglecting other impurities. For both natural gas and ethanol steam reforming systems, the minimum separation exergy translates into less than 1% of the chemical energy of the feed (0.75% and 0.82% for the separators in the natural gas and ethanol systems, respectively). The exergy input for separation may be viewed as the thermo-mechanical exergy of the steam applied for regeneration of the material (e.g., alkaline solution) used to separate the carbon dioxide constituent.

The exergetic efficiency used in this study is defined as

$$\varepsilon = b \left( \frac{\bar{x}_{H_2}}{\bar{x}_F} \right) \quad (8)$$

The energy efficiency is defined as

$$\eta = b \left( \frac{\overline{LHV}_{H_2}}{\overline{LHV}_F} \right) \quad (9)$$

In Equations (8) and (9), the coefficient  $b$  is the number of moles of hydrogen product per mole of the feed, and  $\overline{LHV}$  is the lower heating value in kJ per kmole of the substance. As will be seen later, the exergy efficiency is fairly close to the energy efficiency. This agreement is also demonstrated by another study [13].

Upon rearrangement, Equation (4) can be expressed as

$$\begin{aligned} & \left[ \int (1 - T_o / T_b) \delta \bar{q} \right]_{\text{Endothermic}} + \bar{x}_{\text{elec.}} + \bar{x}_{\text{sep}} + \bar{x}_F + a \bar{x}_{\text{H}_2\text{O}} - b \bar{x}_{\text{H}_2} = \\ & \underline{- \left[ \int (1 - T_o / T_b) \delta \bar{q} \right]_{\text{System Cooling}} + c \bar{x}_{\text{CO}_2} + \bar{x}_{\text{cons.}} > 0} \end{aligned} \quad (10)$$

The underlined term in this equation represents the unused exergy. Considering that the exergy consumed (also referred to as exergy destroyed or irreversibility) within the control volume (CV 1 in Figure 1),  $x_{\text{cons.}}$  is always positive, examination of Equation (9) leads to the following observations:

A lower reformer operating temperature can help reduce the exergy transfer accompanying the heat input and, consequently, reduce the total irreversibility and unused exergy. However, depending on the catalyst type and volume, lowering the operating temperature can also reduce the hydrogen yield and consequently negate the efficiency gains. The concentration of hydrogen in the gaseous mixture leaving the reformer (upstream of the shift reactor) is theoretically influenced by the following second-law based equilibrium criterion.

$$-\frac{\Delta G^\circ}{RT} = \ln K(T) \quad (11)$$

where  $\Delta G^\circ$  is the change in Gibbs function for the reaction and  $K$  is the equilibrium constant at a given reaction temperature. However, the hydrogen concentration determined by Equation (11) is more favorable than what is expected in real-world applications, where the product mixture does not quite reach equilibrium at a given reactor temperature.

The unused exergy departing the system due to the cooling process vanishes as the cooling occurs at a temperature approaching that of the reference environment. It should be noted that a low-temperature cooling would be advantageous only if it is achieved as a result of exhausting all available options for heat recovery, with the lowest possible temperature difference between the heat source and the target media. Otherwise, internal exergy destruction may occur even if the exergy loss associated with the cooling is insignificant.

To improve the hydrogen yield, it is a common practice to supply a higher steam-to-fuel ratio to the reformer than the amount prescribed by the stoichiometric reactions [9, 10, and 11]. Therefore, introducing any excess steam will not only require a higher influx of

energy/exergy for heating but will likely exhibit a higher amount of unused exergy due to the discharge of the excess steam (not shown in Figure 1) – unless it receives a credit for an external use.

For the type of electrolyzer used in this study, the exergy balance (Equation 10) reduces to the following.

$$\bar{x}_{elec.} + \bar{x}_{H_2O} - b' \bar{x}_{H_2} = - \underbrace{\left[ \int (1 - T_o / T_b) \delta \bar{q} \right]}_{System \ Cooling} + c' \bar{x}_{O_2} + \bar{x}_{cons.} > 0 \quad (12)$$

In this equation,  $\bar{x}_{elec.}$  represents the total exergy input, including the electrical exergy (energy) use for the ancillary equipment and  $\bar{x}_{sep}$ . The coefficients  $b'$  and  $c'$  denote the number of moles of hydrogen and oxygen, respectively, in the actual reaction. As before, the underlined term is the unused exergy.

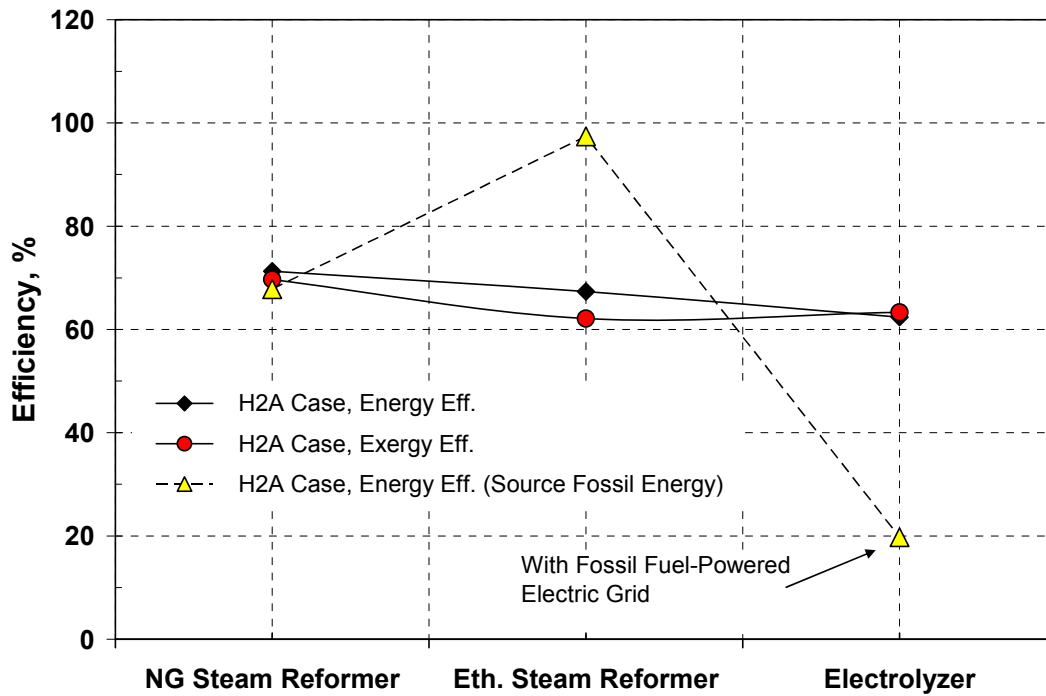
#### 4. Results and Discussion

Figure 3 presents the energy and exergy efficiencies of the three H2A distributed hydrogen production cases: natural gas steam reformer, ethanol steam reformer, and electrolyzer. As seen in this figure, when the efficiencies are determined based on the delivered energy, the energy efficiencies are within about 60% - 70%, with the natural gas reformer being the most efficient. The exergetic efficiencies of the natural gas reformer and electrolyzer are approximately the same as the corresponding energy efficiencies. This congruity can be attributed to:

1. Relatively small thermo-mechanical exergy contribution relative to the chemical exergy of the stream.
2. Closeness of the heating value to chemical exergy of the energy carriers.

For the case of the ethanol reformer, a noticeable, but not significant, difference between the two efficiencies is noted primarily due to a greater difference between the LHV and chemical exergy of liquid ethanol. Close agreements between the two thermodynamic performance indices for other hydrogen technologies have been also demonstrated by Rosen [13].

When the energy efficiency values are determined based on the source energy input, the superiority of the ethanol reformer stands out (Figure 3). This favorable outcome is based on a hydrogen-to-fossil energy ratio of 1.5. Even significantly higher performance can be expected with ethanol produced in state-of-the-art facilities that emphasize use of renewable energy. The electrolysis system using grid electricity is shown to be the least attractive system from the standpoints of fossil fuel consumption and emissions. Even if 50% of the grid electricity were to be supplied by renewable technologies (e.g., wind and PV) on average, the energy efficiency value that only accounts for the fossil energy input would merely increase to about 40%.



**Figure 3. Energy and Exergy Efficiencies.**

Table 1 compares the efficiency results of this study (based on the delivered energy not the source energy) with those of the studies selected from the literature. This table reflects a noticeable scatter among the reported data, which can be attributed to the differences in (1) the operating conditions, (2) the methodologies and assumptions, and/or (3) the technological characteristics of the investigated systems.

Using the approach discussed above, a more detailed exergy analysis of the H2A cases was performed for the three distributed hydrogen production technologies. The results are illustrated in Figure 4. The stacked columns in this figure depict the exergy values of the energy flows entering and leaving the systems (Figures 1 and 2) in terms of MJ per kg of hydrogen produced. The total unused/destroyed exergies within the entire system (CV 1 and CV2 combined) and the burners of the two reformers (CV 2) are also shown.

For the natural gas and ethanol reformers, the relative magnitudes of the flow exergy accompanying the by-product CO<sub>2</sub> and the electrical exergy input are either very small or even negligible (Figure 4). Therefore, inclusion or exclusion of either exergy component in the calculation of the exergetic efficiency will make no significant difference. The exergy value of the heat transfer to the reformer (the first term on the right side of Equation (6) is estimated using a constant operating temperature of 850°C (applied in the H2A model) as the boundary temperature. The amount of heat is determined based on the fraction of the total fuel input (feed plus combustion fuel) required for the burner, as previously discussed – 0.32 for the natural gas reformer and 0.28 for the ethanol. For the steam reformers, the unused/destroyed exergy equals the difference between the heat

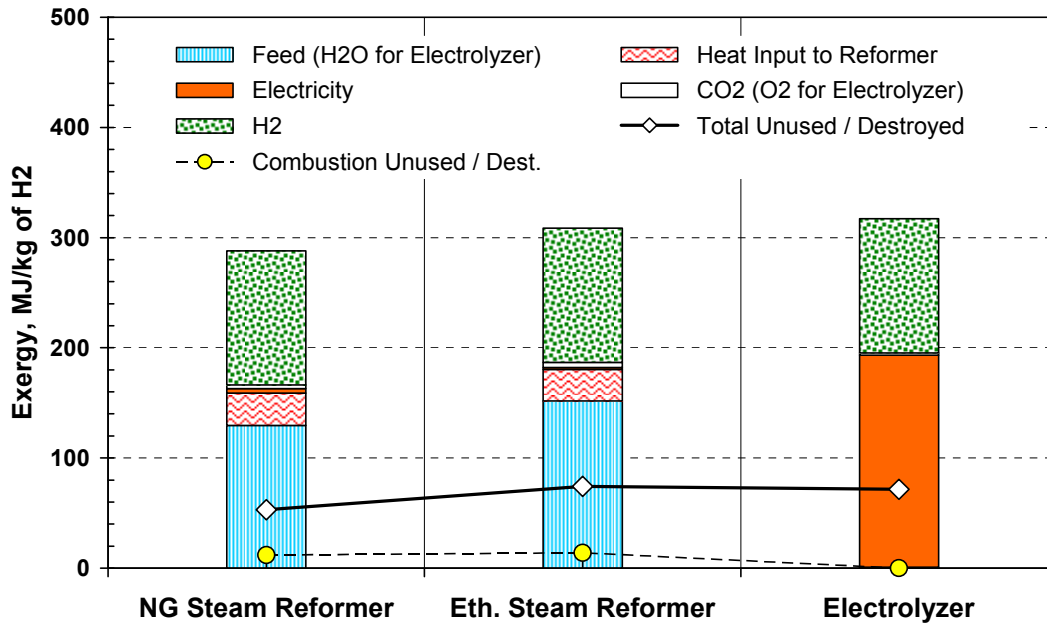


transfer exergy and the chemical exergy of the combustion fuel. The variation of the unused/destroyed exergies with the technology type is consistent with the exergetic efficiency trend depicted in Figure 3.

**Table 1. Comparison of efficiency results with selected data from the literature.**

Source	Natural Gas Steam Reformer		Ethanol Steam Reformer		Electrolyzer	
	$\eta$ (%)	$\varepsilon$ (%)	$\eta$ (%)	$\varepsilon$ (%)	$\eta$ (%)	$\varepsilon$ (%)
Current Study <sup>(1)</sup>	71	70	67	62	62	63
Ref. [14] <sup>(2)</sup>	73-75	---	---	---	64-70	---
Ref. [13]	86	78	---	---	30-49	26-41
Ref. [15]	---	---	---	---	59	---
Ref. [12] <sup>(3)</sup>	---	60-67	---	---	---	---

1. Determined using the results of the latest H2A Production case models based on the current state-of-the-art technologies.
2. Includes the net output of by-product (e.g., steam). The ranges are provided based on literature review.
3. The reported range is for reformed product temperatures of 1100 K and higher.



**Figure 4. Exergy balance for hydrogen production systems.**

For the electrolyzer, the total electrical exergy (energy) input is the equivalent of the combined exergy (energy) of the feed, combustion fuel, and electricity used in the reformers. Given the second-law constraint on the fossil-to-electricity efficiency, the overall exergetic performance of the electrolyzer (fossil fuel to hydrogen conversion) is significantly less attractive unless renewable sources of energy are major contributors to

the electrical grid. When fully integrated with an on-site renewable power generator (e.g., wind energy or PV), the need for fossil-source energy diminishes for the electrolyzer. Regardless of the system, steam reformer or electrolyzer, any reduction in the fossil fuel consumption proportionately reduces the greenhouse gas (GHG) emissions.

Figure 5 provides the cost data for hydrogen production using the selected H2A cases. The stacked columns demonstrate the relative contributions of the main cost categories: energy (feed and combustion fuel), capital, and operating and maintenance. The total hydrogen cost represented by the solid line (Figure 5) is normalized based on the unit cost of hydrogen produced by the natural gas reformer. Clearly, the total cost of energy is dominant: 62% for the natural gas reformer, 77% for the ethanol reformer, and 58% for the electrolyzer. It should be pointed out that the higher energy cost of the ethanol reformer partly stems from the capital cost associated with the ethanol production. The capital cost is the second in significance – less than 30%. In contrast, the data compiled by a European study [14] on various hydrogen production technologies, which did not include ethanol reforming, suggest a different cost composition for small-scale natural gas reformers and electrolyzers. In that study, the operations and maintenance (O&M) cost has a considerably greater share of the total cost. Figure 5 indicates that hydrogen production by the electrolyzer is the most expensive system, followed by the ethanol reformer.

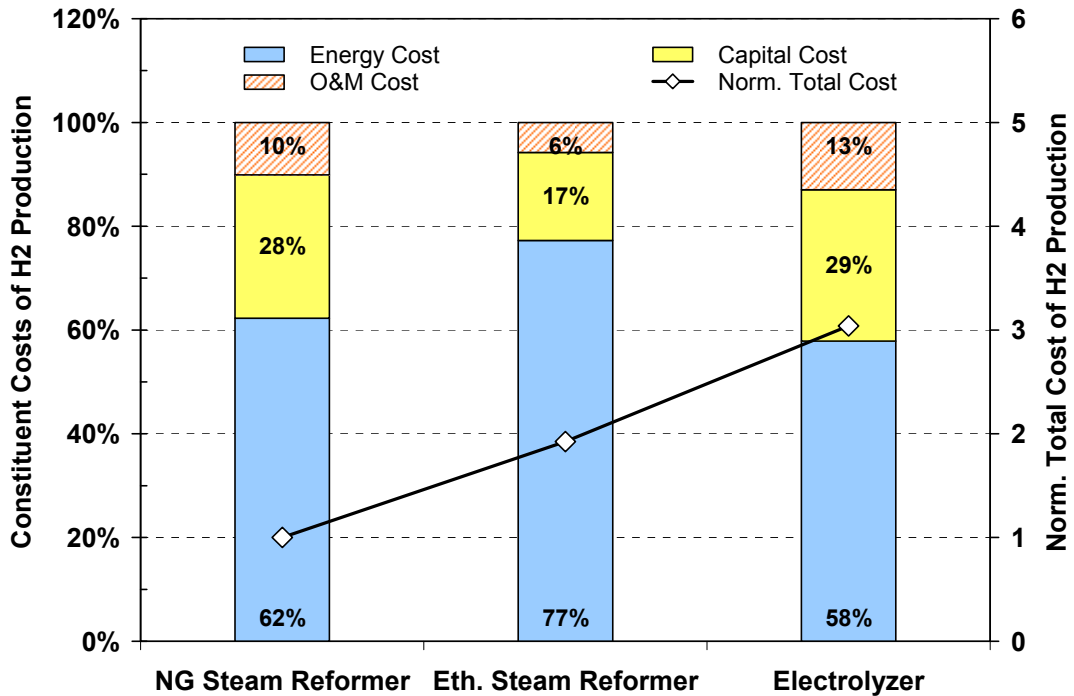


Figure 5. Costs of hydrogen production.

The confluence of the thermodynamic and economic results (Figures 4 and 5) has important implications. On the one hand, Figure 5 suggests that there may be an opportunity for a substantial reduction in the hydrogen cost by curtailing the dominant energy (feed, fuel, and electricity) share of the total cost. On the other hand, the unused/destroyed portion of the total exergy input (Figure 4) limits this perceived opportunity. For example, for the natural gas reformer, assuming that the capital and O&M costs remain intact, the maximum theoretically possible reduction in energy use translates into a hydrogen cost reduction of about 20%. However, in real-world applications, the energy-related savings cannot be this high because of insurmountable challenges in eliminating the exergy destruction and in recovering low-grade heat loss. Similar analyses, in concert with inputs from real-world experiences, are imperative for realistically predicting the cost-reduction potentials for the future infrastructural systems.

## 5. Conclusions

In this paper, economic and thermodynamic analyses were conducted for three distributed hydrogen production systems – a natural gas steam reformer, an ethanol steam reformer, and an electrolyzer. The economic evaluation was based on the cost data obtained from the H<sub>2</sub>A Production case studies. For the thermodynamic analysis, the attention was devoted to both energetic and exergetic performance of the selected systems. The impact of the intermediate process that occurs between the source and the point of delivery was addressed from the thermodynamic standpoint for the required inputs of energy and feedstock. The relative contributions of the key cost and exergy constituents were elaborated.

The first- and second-law efficiencies for the gas reformers were either virtually equal or slightly different, owing to the slight difference between the corresponding heating values and chemical exergies of the feed (natural gas) and hydrogen. For the ethanol reformer, the difference between the two performance indices was noticeable, but not very significant. Roughly equal energy and exergy efficiencies were also observed with the electrolyzer. Overall, the results from the energy and exergy analyses were rather similar. The only major difference between the two approaches pertains to the thermal interaction between the burner and the reforming process, as the exergy accompanying the heat transfer is influenced by the operating reactor temperature. However, this exergy component has a limited role in the overall system performance.

For the investigated cases, the energy cost has a greater share than the sum of capital and non-energy operating costs, particularly for the case of the ethanol reformer. However, any predictive analyses attempting to project energy cost reduction through efficiency improvements are subject to the ineluctable laws of thermodynamics, as demonstrated in this study.

## 6. Nomenclature

$a, b, c$	Molar coefficients of substances in chemical reaction, kmol / kmol of fuel
$\bar{h}$	Specific enthalpy, kJ/kmol of fuel
$\overline{HHV}$	Higher heating value, kJ/kmol of fuel
$K$	Equilibrium constant
$\overline{LHV}$	Lower heating value, kJ/kmol of fuel
$P$	Pressure, kPa
$\delta \bar{q}$	Differential heat transfer, kJ/kmol of fuel
$\bar{R}$	Universal gas constant, 8.314 kJ/kmol.K
$\bar{s}$	Specific entropy, kJ/kmol.K
$T$	Temperature, K
$\bar{x}$	Flow exergy, kJ/kmol
$\Delta G^o$	Change in Gibbs function, kJ/kmol
$\varepsilon$	Exergetic (second-law) efficiency
$\eta$	Energetic (first-law) efficiency

### Subscripts

$b$	Boundary
$F$	Fuel / feed
$m$	Mixture
$o$	Reference environment
$sep.$	Associated with separation of mixed gases

### Superscripts

$ch$	Chemical
$thm$	Thermo-mechanical

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