



THERMODYNAMIC COMPARISON OF HYDROGEN PRODUCTION PROCESSES

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Abstract—The results are reported of comparisons based on energy and exergy analyses of a wide range of hydrogen production processes, including processes which are hydrocarbon-based (steam-methane reforming and coal gasification), non-hydrocarbon-based (water electrolysis and thermochemical water decomposition) and integrated (steam-methane reforming linked to the non-hydrocarbon-based processes). A version of the Aspen Plus process-simulation computer code, previously enhanced by the author for exergy analysis, is used in the analyses. Overall efficiencies are determined to range widely, from 21 to 86% for energy efficiencies, and from 19 to 83% for exergy efficiencies. The losses in all processes are found to exhibit many common factors. Energy losses associated with emissions account for 100% of the total energy losses, while exergy losses associated with emissions account for 4–10% of the total exergy losses. The remaining exergy losses are associated with internal consumptions. It is anticipated that the results will prove useful to those involved in the improvement of existing and design of future hydrogen production processes. Copyright © 1996 International Association for Hydrogen Energy.

INTRODUCTION

Hydrogen is used as a fuel, and as a feed for producing other fuels and commodities. In the future, the role of hydrogen may become more important, as some researchers suggest that the world's energy systems may undergo a transition to an era in which the main energy carriers are hydrogen and electricity [1–3]. Since the production of hydrogen had been identified in the past as in need of substantial R&D [4, 5], the author established a research program in the 1980s to assess and compare, using energy and exergy analyses, the thermodynamic performance of many hydrogen production processes. The work reported here forms part of that program, and compares the results of many of the author's previous investigations [6–20] of the energy- and exergy-based performance for various hydrogen production processes. The intent of this publication is to develop a better understanding of the thermodynamic characteristics of hydrogen production processes, thereby aiding in optimization and improvement efforts, and to provide information that assists in decision making regarding alternative hydrogen-production processes.

The main processes for hydrogen production include steam-methane reforming (SMR), catalytic decomposition natural gas, partial oxidation of heavy oil, coal gasification, water electrolysis, thermochemical water decomposition, and photo-chemical, -electrochemical and -biological processes [1–5]. The first four processes

are based on fossil fuels. SMR, coal gasification and water electrolysis are the most important industrial processes for hydrogen production today. The hydrogen production processes considered in this study are listed in Table 1 by category (hydrocarbon-based, non-hydrocarbon-based and integrated).

Table 1. Hydrogen production process considered, by category

Category	Process
Hydrocarbon-based	Steam-methane reforming (SMR) Coal gasification
Non-hydrocarbon-based	Current-technology water electrolysis Thermochemical water decomposition
Integrated	SMR/Current-technology water electrolysis SMR/Advanced-technology water electrolysis SMR/Thermochemical water decomposition

In Table 1:

- only processes that are significant and for which substantial and reliable design and operating data are available are included;
- current and advanced technologies for water electrolysis are considered; and

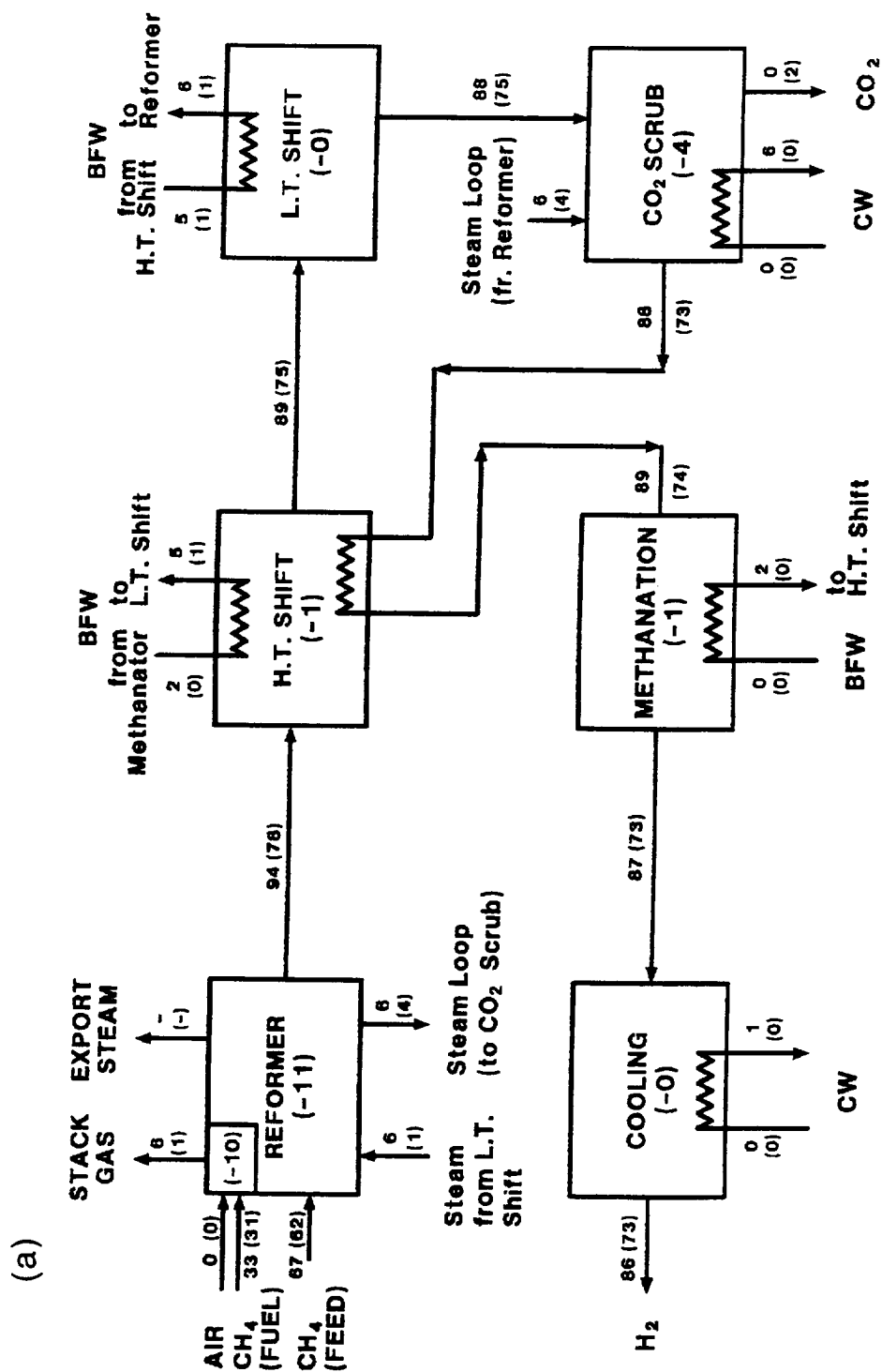


Fig. 1(a).

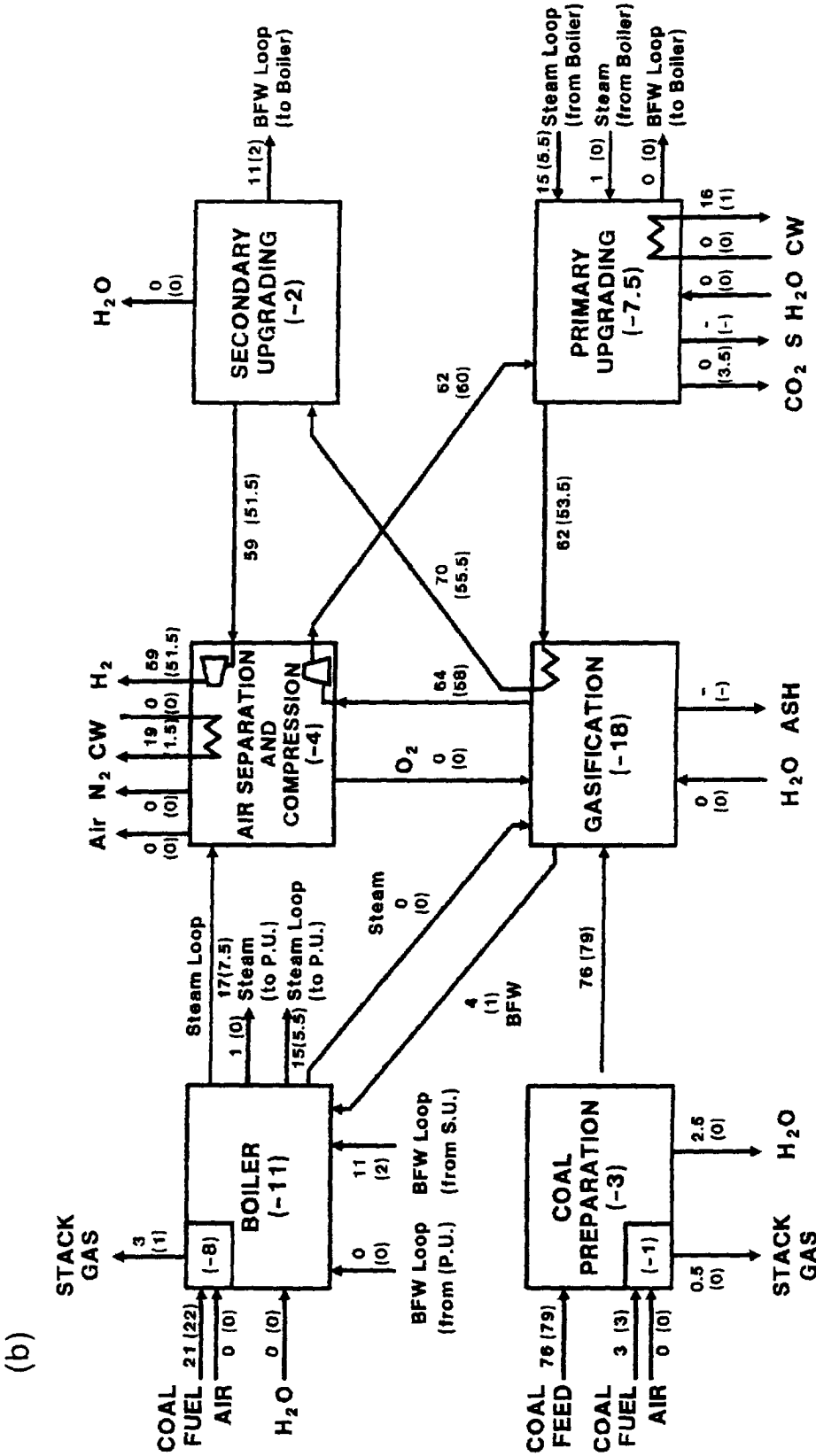


Fig. 1(b).

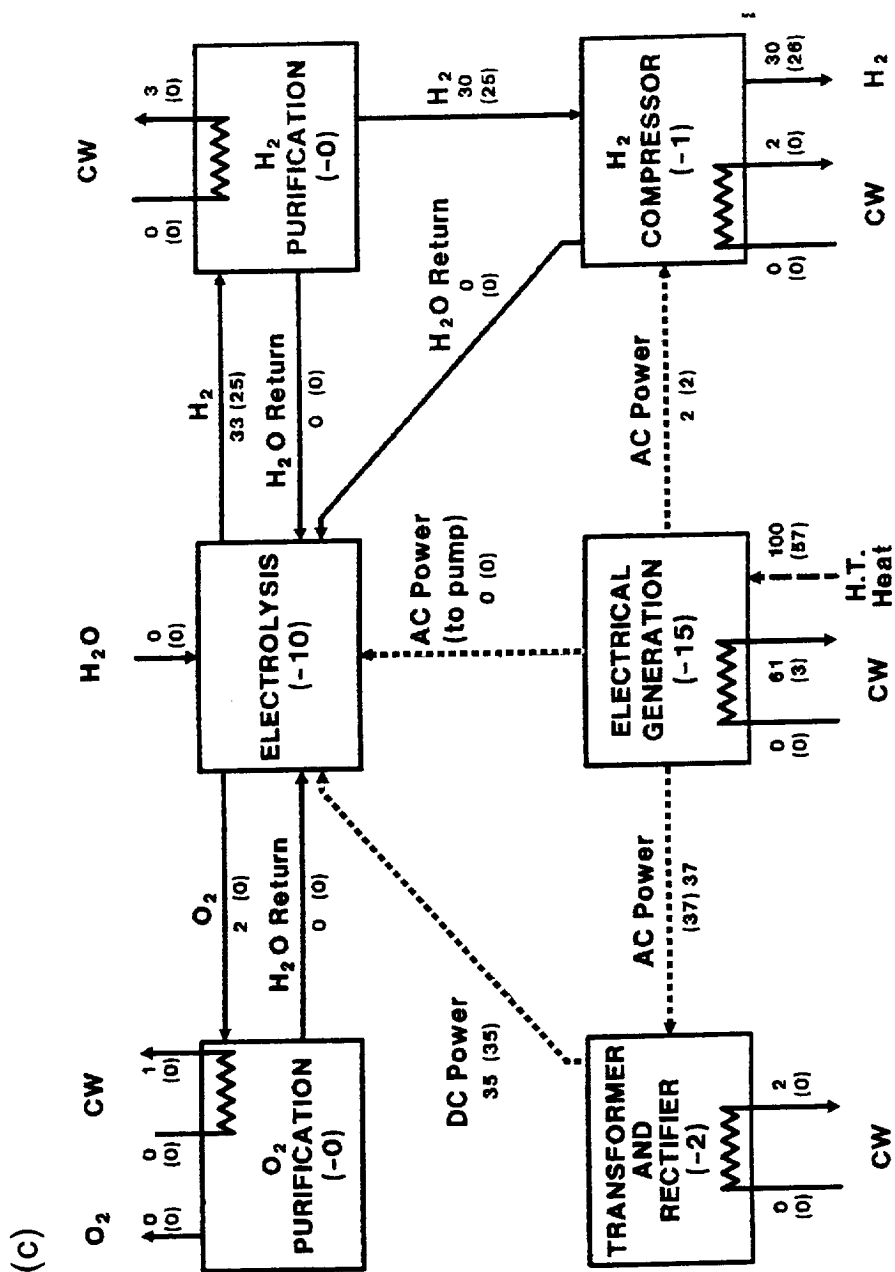


Fig. 1(c).

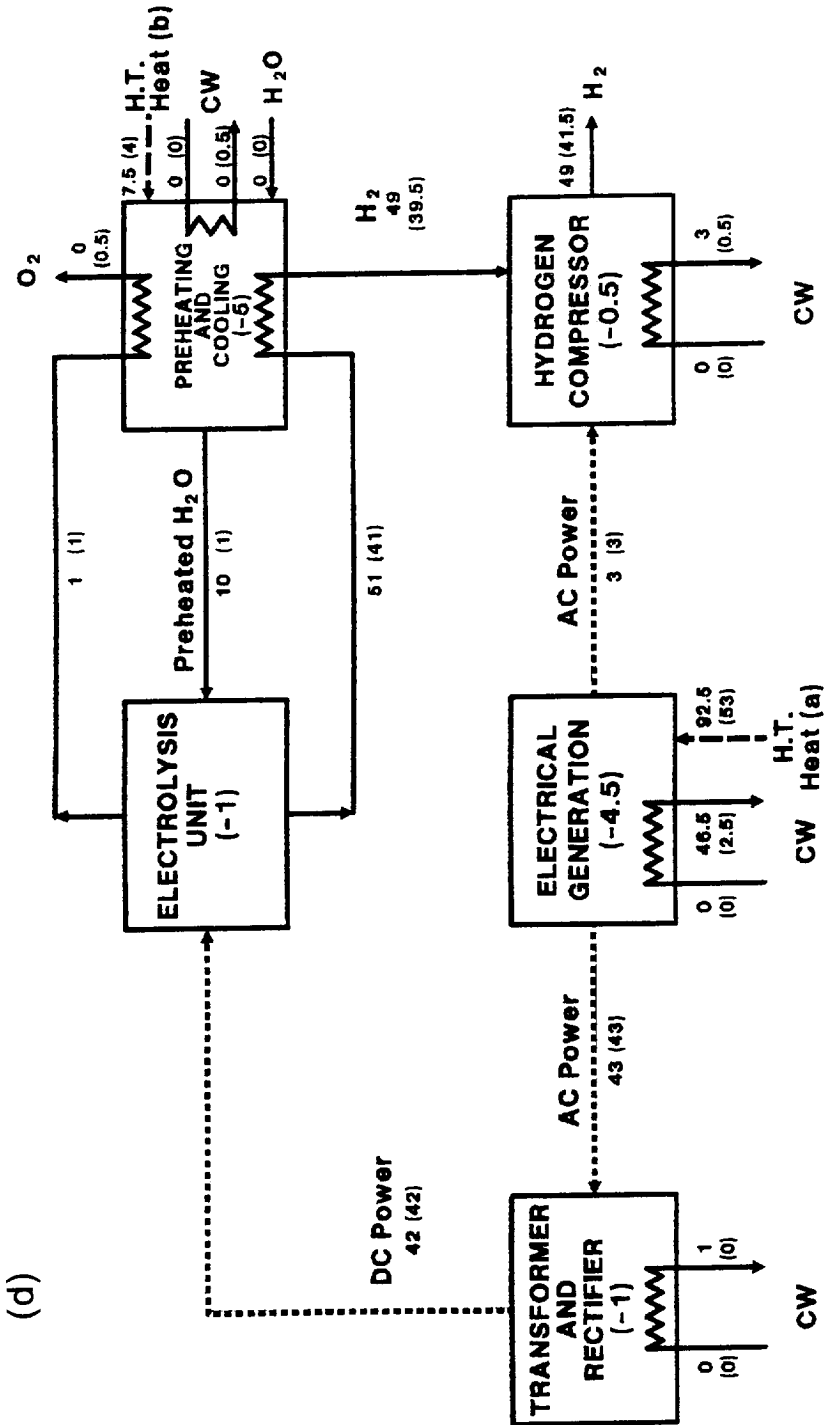


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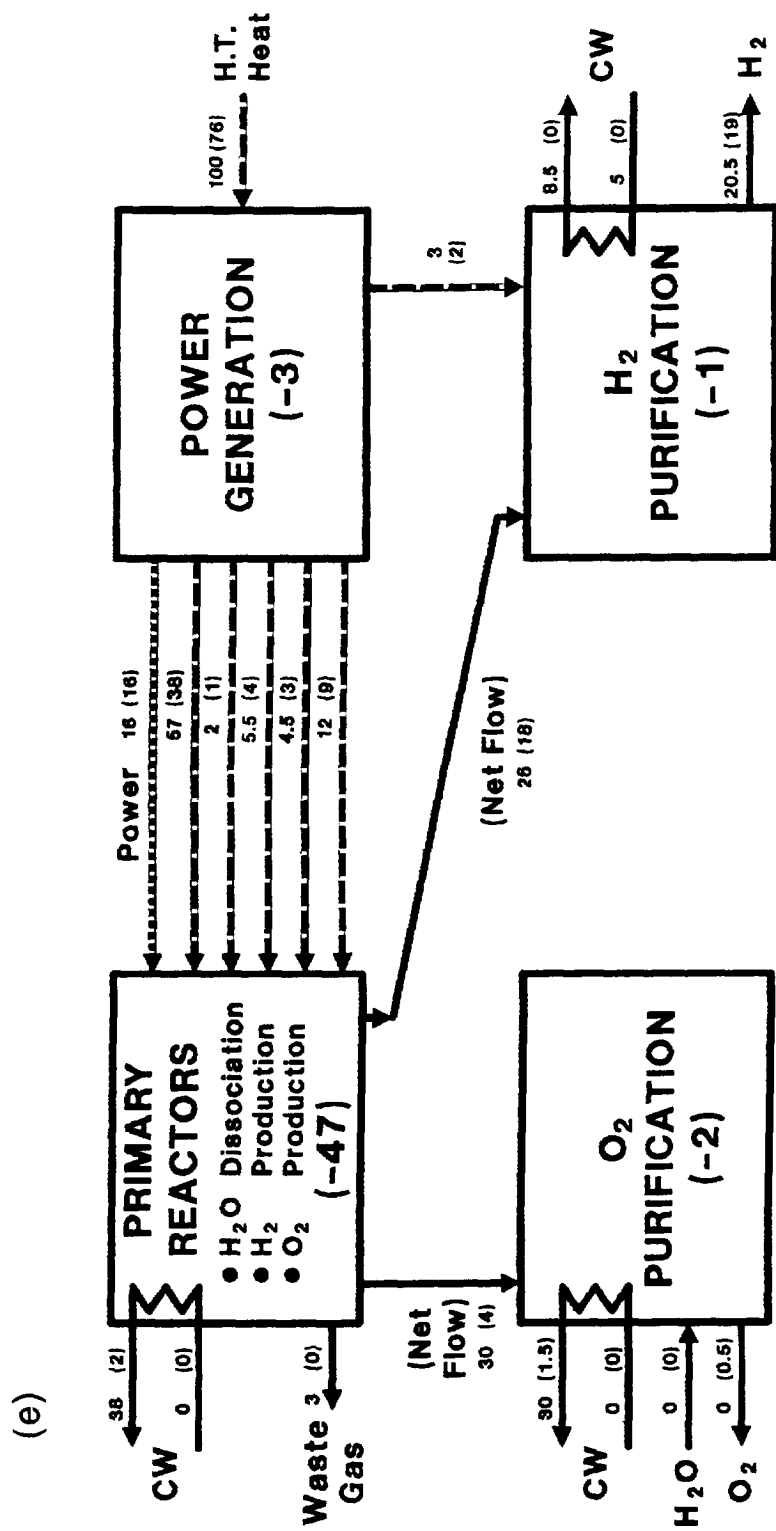


Fig. 1(e).

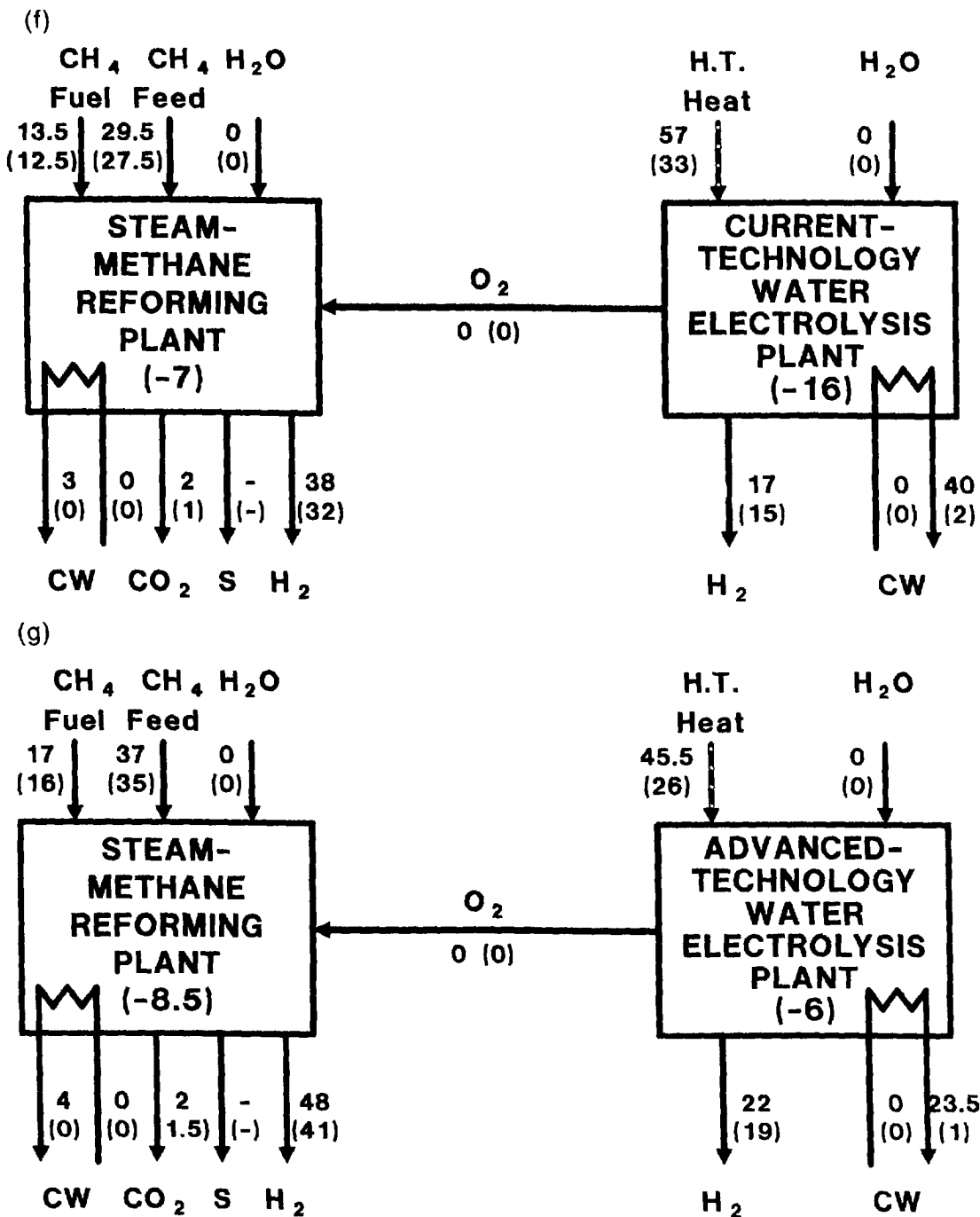


Fig. 1. (f), (g).

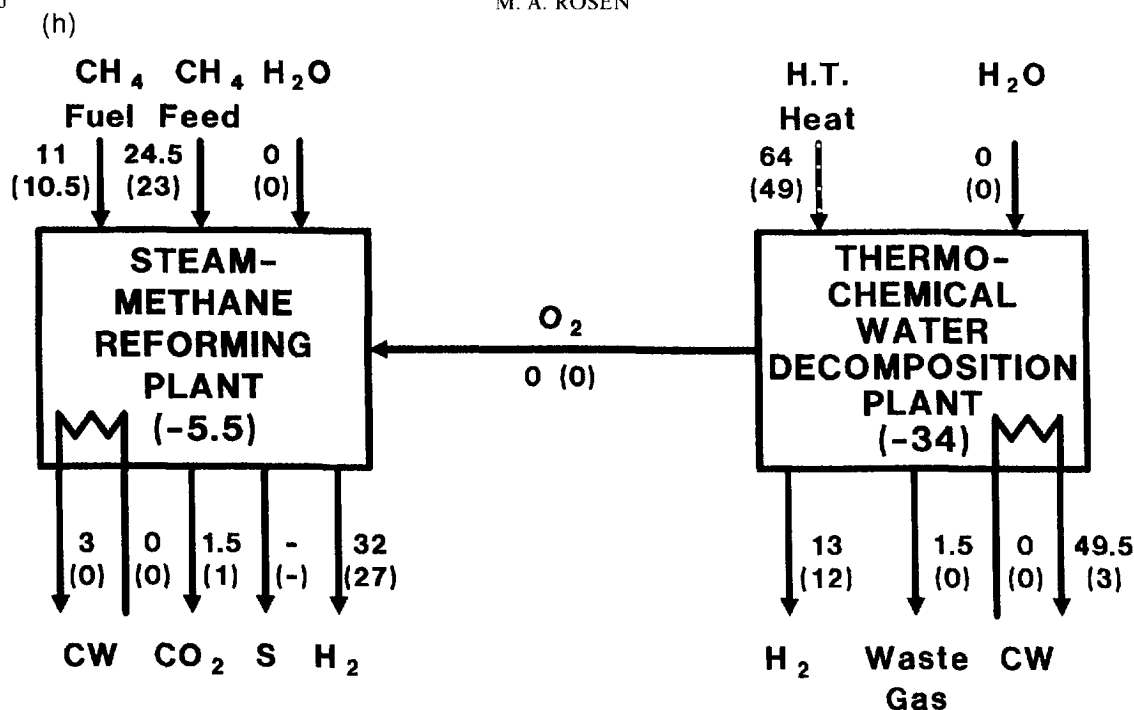


Fig. 1. Simplified process diagrams for hydrogen production indicating flow rates of energy (values not in parentheses) and exergy (positive values in parentheses) for streams, and exergy consumption rates (negative values in parentheses) for devices. The portions of exergy consumption due to the combustion reaction are indicated in small squares in the appropriate sections of (a) and (b). All values are normalized so that 100 U of energy enter each overall process. Descriptions of input and output streams for the overall process are given in upper case. (a) SMR; (b) coal gasification; (c) current-technology water electrolysis; (d) advanced-technology water electrolysis; (e) thermochemical water decomposition; (f) SMR/current-technology water electrolysis (g) SMR/advanced-technology water electrolysis; (h) SMR/thermochemical water decomposition

(iii) the integrated processes use by-product oxygen from water electrolysis or thermochemical water decomposition to enhance combustion in the SMR furnace.

Energy and exergy analyses [21–23] are used here to perform thermodynamic performance comparisons. Energy analysis is based on the first law of thermodynamics, which is concerned with the conservation of energy. Exergy analysis is based on the second law, and generally allows the causes and locations of process inefficiencies to be better pinpointed than does energy analysis, and efficiencies to be more rationally evaluated. Many researchers (e.g. [21–23]) propose that the thermodynamic performance of a process is best evaluated using exergy analysis. Exergy is the maximum amount of work which can be produced by a stream or system as it is brought into equilibrium with a reference environment, and can be thought of as a measure of the usefulness or quality of energy. Exergy is consumed during real processes, and conserved during ideal processes. The exergy consumption during a process is proportional to the entropy created due to process irreversibilities.

PROCESS DESCRIPTIONS

Detailed process diagrams for the production processes selected for analysis are presented in Fig. 1, and corresponding process data and descriptions in Table 2

(broken down for each process according to the individual process sections shown in Fig. 1).

Table 2. Descriptions of the hydrogen production processes considered

Steam-methane reforming (SMR) [36 (pp. 42–44), 37–39]

Reforming and steam generation. CH₄ feed is desulphurized and mixed with superheated steam. Reforming occurs and raw gas exits at 370°C and 3.5 MPa. Steam is generated in the boiler for compression and CO₂ stripping. Required process heat is supplied by combusting CH₄ fuel.

High-temperature shift. 94% of the CO in the raw gas is shifted over a high-temperature catalyst. Gas exits at 220°C and preheats incoming boiler and methanator feeds.

Low-temperature shift. 83% of the remaining CO in the raw gas is shifted over a low-temperature catalyst. Gas exits at 150°C and preheats incoming feedwater.

Compression and carbon-dioxide removal. Raw gas is compressed to 3.5 MPa with steam-turbine-driven centrifugal compressors. CO₂ is removed using the monoethanolamine process, leaving the CO₂ content of the raw gas at 0.1% by weight. Steam supplies energy (821 Btu/lb CO₂ recovered) to the CO₂ stripper.

Methanation. Steam preheats the methanator feed to 350°C and 2.4 MPa, and methanation occurs over a catalyst.

Cooling. Gas preheats feedwater and is cooled to 25°C with cooling water. Water is separated and the product (97 wt% H₂) exits at 25°C and 2.4 MPa.

Coal gasification (based on the Koppers-Totzek process)
[36 (pp. 46–50), 37–39]

Coal preparation. Raw coal (22% moisture) is input as feed and fuel. Feed coal is dried to 2% moisture and pulverized.

Steam generation. Steam is generated by combusting boiler-fuel coal.

Compression and air separation. Raw gas is compressed from 20 to 700 psig, and product gas from 700 to 1000 psig. An air separation plant produces O_2 . Superheated steam drives the turbines.

Coal gasification. Feed coal is gasified in the presence of O_2 and low-pressure steam. 50% of the molten slag is removed from the raw gas (at 2730°F and 20 psi), and water is added to quench the gas to 100°F. Superheated steam at 1200 psig and 900°F is generated from the raw gas. Raw gas (180°F) is cleaned with a venturi scrubber and electrostatic precipitator.

Primary upgrading of raw gas. H_2S is removed with the Rectisol system. Steam and quench water is added. CO concentration is reduced from 59% to 3% by shifting the raw gas. Raw gas (at 370°F and 700 psig) preheats feedwater, and is cooled to 115°F with cooling water. CO_2 is removed with the monoethanolamine process.

Secondary upgrading of raw gas. Raw gas is preheated to 550°F and the CO content reduced to 0.1% by methanation. Raw gas is cooled from 900 to 125°F by heat recovery and water cooling, and dried to 7 lb-water/10⁶-scf-gas. Product gas (93% H_2) is compressed to 1000 psig.

Current-technology water electrolysis [36 (pp. 162–171), 37–39]

Electrical generation. Electricity is generated from thermal energy at 39% efficiency. Some electricity is delivered to the compressor, and most to the transformer.

Transformation and rectification. Electricity is converted from AC to DC at 97% efficiency.

Electrolysis. Feed water is purified and mixed with recycled water from the coolers and compressors in the electrolyte circulation-loop. Water is decomposed in the electrolysis unit, and raw gases exit at 359 K and 0.101 MPa.

Hydrogen cooling. H_2 stream is cooled to 308 K with cooling water. Moisture condenses and is recycled.

Oxygen cooling. O_2 stream is cooled to 308 K with cooling water. Moisture condenses and is recycled.

Hydrogen compression. H_2 is compressed in a 4-stage compressor with intercooling to 308 K. Moisture condenses and is recycled. Product gas (99.5 wt% H_2) exits at 10.3 MPa and 308 K.

Advanced-technology water electrolysis [36 (pp. 162–171), 37–39]

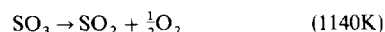
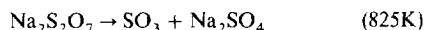
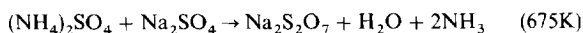
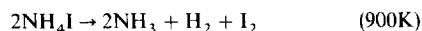
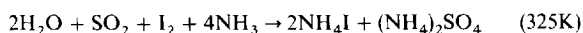
Same as current-technology water electrolysis except: (i) electricity is generated at 50% efficiency; (ii) added to the H_2 and O_2 cooling sections, which are combined, is feedwater preheating, where feedwater is purified and heated, with hot gases from electrolysis (with 80% heat recovery) and direct heat input, to 800 K and 0.101 MPa; (iii) the electrolysis step involves no recycle flow; and (iv) the product gas is 100% H_2 .

Thermochemical water decomposition (based on the Ispra Mark-10 cycle) [36 (pp. 254–282), 37–39]

Thermal and electrical generation. Helium delivers thermal energy from a 'high-temperature gas-cooled reactor' at 1255 K and returns at 773 K. Electricity is generated at 80% of the

thermodynamic ideal efficiency. Electricity is used in the primary reactors, and the remainder of the heat is delivered over a range of temperatures to the other units.

Primary thermochemical-water-decomposition reactions. 5 principal reactions for which the net reaction is water decomposition occur over a range of temperatures:



17% of the water in the cycle decomposes, and a waste gas exits at 325 K and 0.101 MPa.

Hydrogen purification. Helium (entering at 815 K and exiting at 798 K) delivers thermal energy for purification. Product gas (99.9 wt% H_2) exits at 300 K and 0.210 MPa.

Oxygen purification. O_2 exits at 300 K and 5.00 MPa, and feedwater enters at 290 K and 0.101 MPa.

SMR/current-technology water electrolysis

SMR. Same as SMR above, except CH_4 fuel is combusted in the reformer furnace in the stoichiometric amount of pure O_2 (from the linked non-hydrocarbon-based process).

Current-technology water electrolysis. Same as current-technology water electrolysis above, except plant size is scaled so the O_2 flow rate exactly matches the requirement of the reformer furnace of the linked SMR plant.

SMR/advanced-technology water electrolysis

Same as SMR/current-technology water electrolysis, except with advanced in place of current-technology water electrolysis.

SMR/thermochemical water decomposition

Same as SMR/current-technology water electrolysis, except with thermochemical water decomposition in place of current-technology water electrolysis.

APPROACH

Analyses and comparisons are performed using a computer code developed by enhancing a state-of-the-art process simulator, Aspen Plus, for exergy analysis [8]. A previously defined reference-environment model [21, 22] is used in the evaluation of energy and exergy quantities, but with a reference-environment temperature (T_0) of 15°C (the approximate mean temperature of the lake cooling water). The reference-environment pressure (P_0) is taken to be 1 atm, and the chemical composition is taken to consist of air saturated with water vapor, and the following condensed phases at 25°C and 1 atm: water (H_2O), gypsum ($CaSO_4 \cdot 2H_2O$), and limestone ($CaCO_3$). In addition to properties in Aspen Plus data banks, which include steam properties based on the 1967 ASME steam tables, base enthalpy and chemical exergy values reported elsewhere [21, 22] are used. The base enthalpy is the enthalpy of a component (at T_0 and P_0) relative to the stable components of the environment (at T_0 and P_0). The base enthalpy of a fuel is equal to the enthalpy change

in forming the fuel from the components of the environment (the same environment used in exergy calculations). A compound which exists as a stable component of the reference environment is defined to have an enthalpy of zero T_0 and P_0 .

To simplify the simulations and comparisons:

- (i) combustion is assumed to occur in 40% excess air;
- (ii) hydrocarbons are considered sulphur free; and
- (iii) natural gas is modelled as pure methane, coal as pure carbon, and air on a volume basis as 79% nitrogen and 21% oxygen.

For processes in which high-temperature heat is an input (i.e. water electrolysis, thermochemical water decomposition and the integrated processes), analyses consider three main inputs:

- (i) electrical power and process heat;
- (ii) heat, and
- (iii) a hypothetical heat source.

A process includes heat and electricity generation steps in the third case, and an electricity generation step in the second. The hypothetical heat source is intended to be an environmental resource. The existence of a heat source is hypothesized so that processes in which some inputs (e.g. high-temperature heat) are not environmental resources can be compared with processes in which all inputs are environmental resources. The exergy of a hypothetical heat source is considered equal to its energy (an approximately valid assumption for many fossil fuels).

RESULTS

Energy and exergy efficiencies, determined as the ratio of the energy (or exergy) of product(s) to the energy (or exergy) of input(s), are presented for the overall processes in Table 3. By-product credits are not allowed, although no significant change is observed in the efficiencies if by-product credits are allowed. The overall efficiencies range widely, from 21 to 86% for energy efficiencies, and from 19 to 83% for exergy efficiencies. Simplified diagrams containing normalized energy and exergy flow rates and exergy consumption rates are presented in Fig. 1 for each process. The exergy consumption rates are further broken down into internal (consumptions) and external (emissions) in Table 4. Overall energy and exergy balances are shown in Fig. 2 [with Figs. 2(c–h) considering hypothetical heat sources as the main inputs].

More detailed results are reported by the author elsewhere for the hydrogen production processes considered: SMR [6–9], coal gasification [9–12], current-technology water electrolysis [9, 13–17], advanced-technology water electrolysis [9, 16–18], thermochemical water decomposition [9, 17, 19], and the integrated processes [20].

The validity of the results is verified by comparing them with the literature for similar processes. Several energy analyses, but only a few exergy analyses, of the processes have been performed. Exergy analyses of the following

Table 3. Energy and exergy efficiencies for the hydrogen production processes considered

Process	Efficiency (%)	
	Energy	Exergy
<i>SMR</i>	86	78
<i>Coal gasification</i>	59	49
<i>Current-technology water electrolysis</i>		
From electricity and process heat	77	67
From heat	30	46
From hypothetical heat source	30	26
<i>Advanced-technology water electrolysis</i>		
From electricity and process heat	92	83
From heat	49	73
From hypothetical heat source	49	41
<i>Thermochemical water decomposition</i>		
From electricity and process heat	21	26
From heat	21	25
From hypothetical heat source	21	19
<i>SMR/current-technology water electrolysis</i>		
From electricity and process heat	84	76
From heat	55	65
From hypothetical heat source	55	48
<i>SMR/advanced-technology water electrolysis</i>		
From electricity and process heat	89	81
From heat	70	78
From hypothetical heat source	70	62
<i>SMR/thermochemical water decomposition</i>		
From electricity and process heat	45	48
From heat	45	47
From hypothetical heat source	45	40

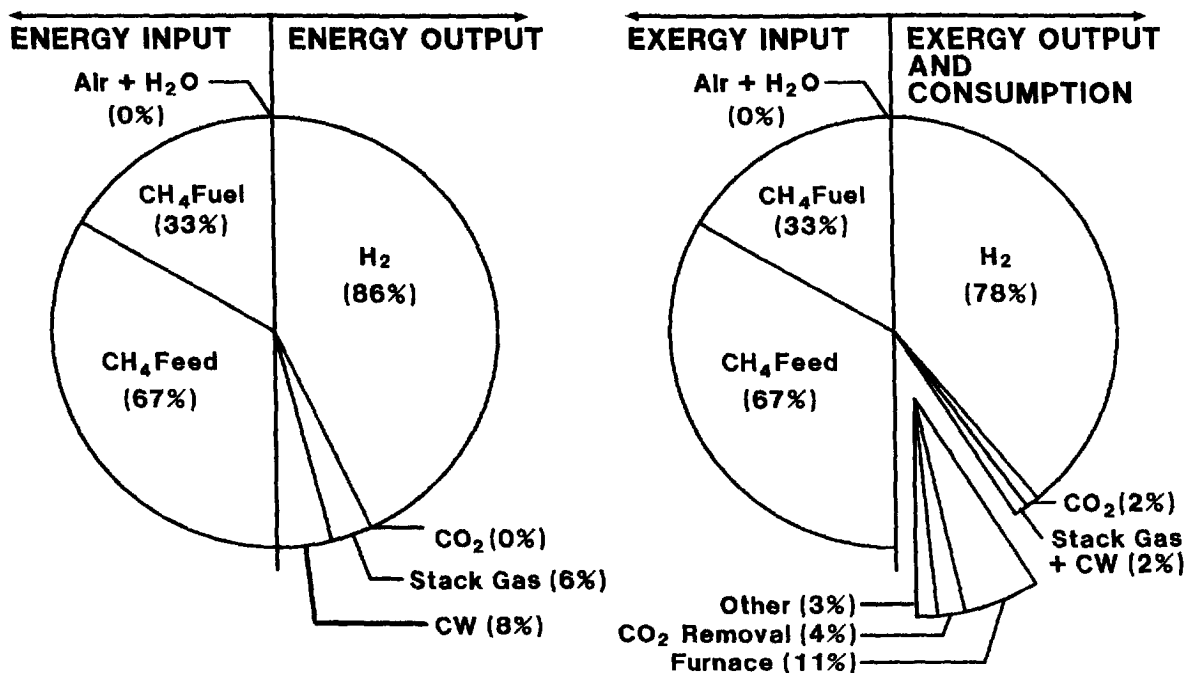
processes are used for validation: combustion [24]; catalytic reforming [25]; coal conversion [26–32]; and electrolytic and thermochemical hydrogen production [33–35]. For each process, the results of energy and exergy analyses obtained here are within the range of and in broad agreement with the results in the literature.

Table 4. Breakdown (in %) of exergy losses for the hydrogen production processes considered*

Process	External	Internal
SMR	9	91
Coal gasification	6	94
Current-technology water electrolysis	4	96
Advanced-technology water electrolysis	4	96
Thermochemical water decomposition	5	95
SMR/current technology water electrolysis	4	96
SMR/advanced-technology water electrolysis	4	96
SMR/thermochemical water decomposition	6	94

* Heat is considered the driving input in the last six cases, and the breakdown is provided as a percentage of the total exergy loss.

(a)



(b)

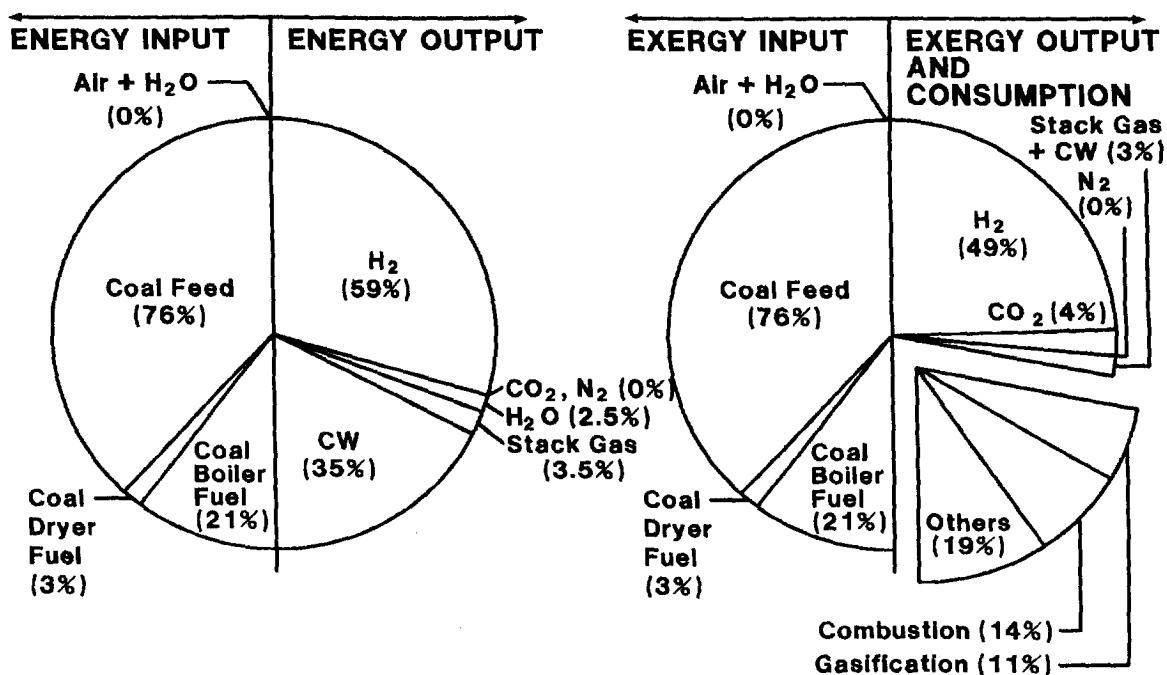
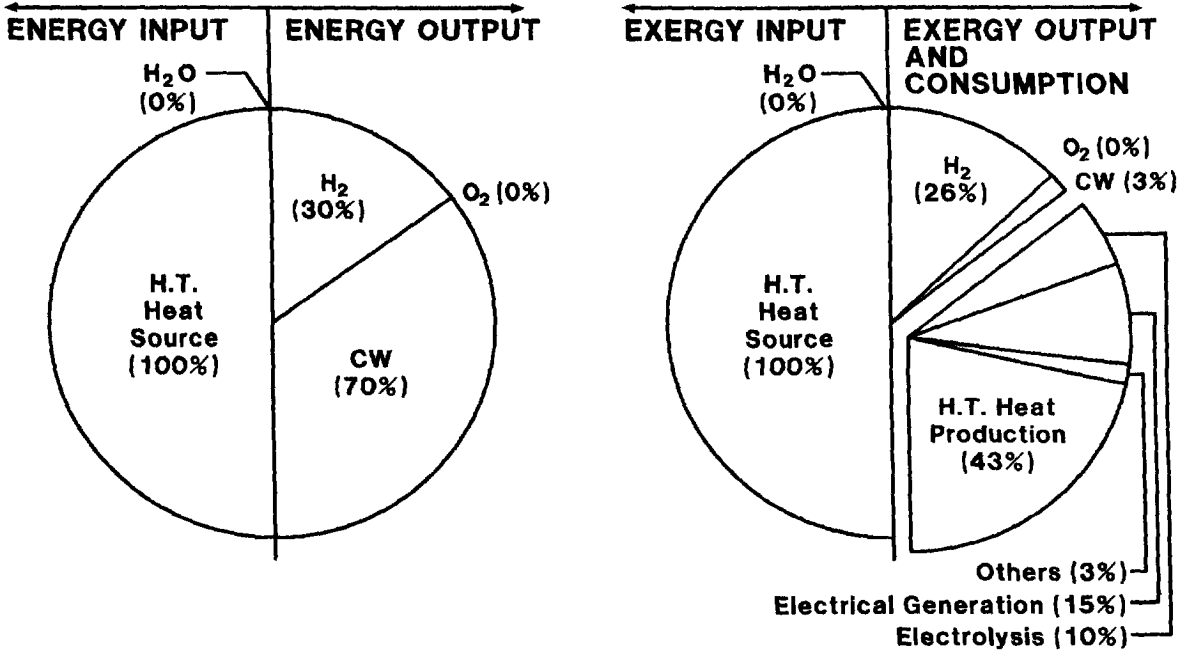


Fig. 2. Overall energy and exergy balances for hydrogen production. The left and right halves of the energy balances represent respectively energy inputs and energy outputs. The left and right halves of the exergy balances represent respectively exergy inputs and exergy outputs and consumptions (exploded section of balance). A source of high-temperature heat is considered as a process input where applicable. (a) SMR; (b) coal gasification.

(c)



(d)

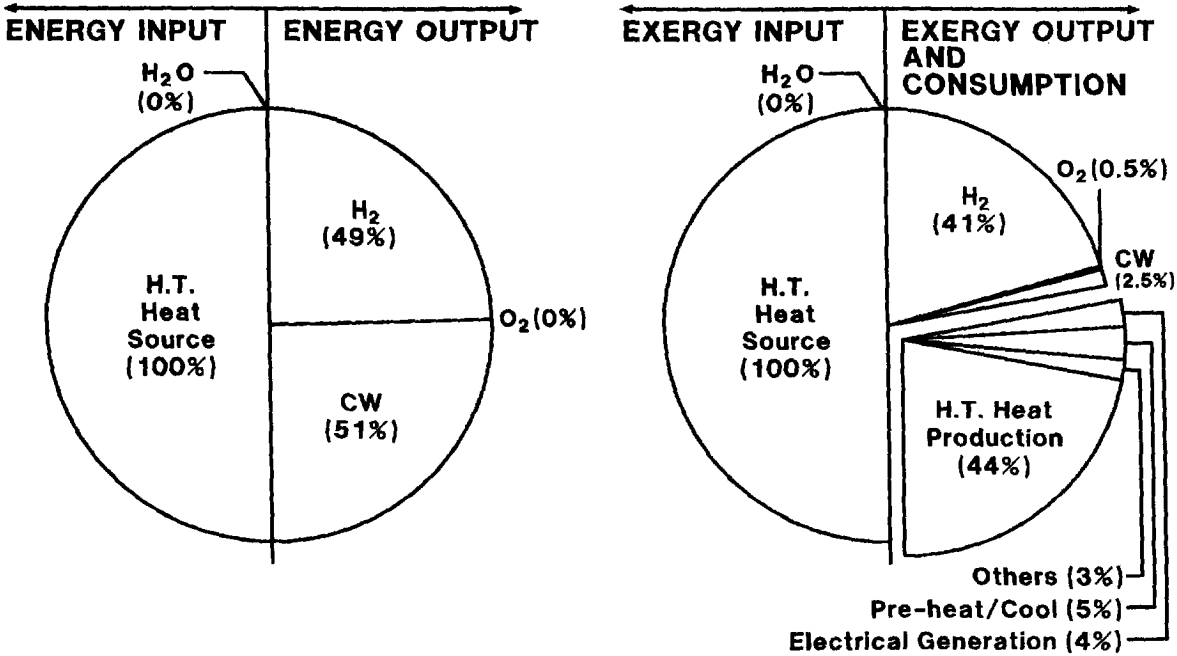
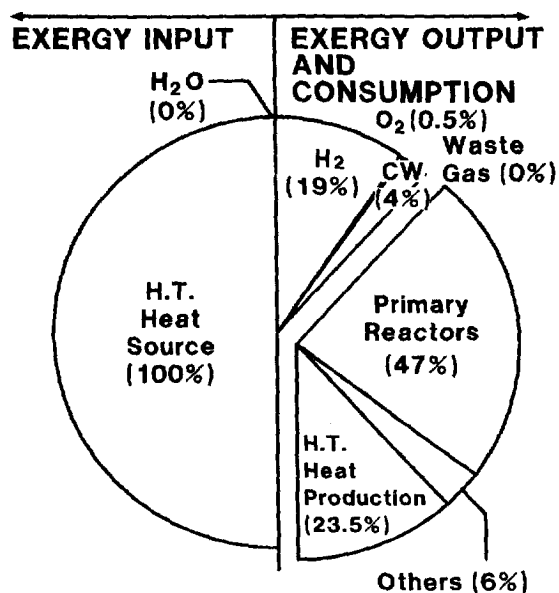
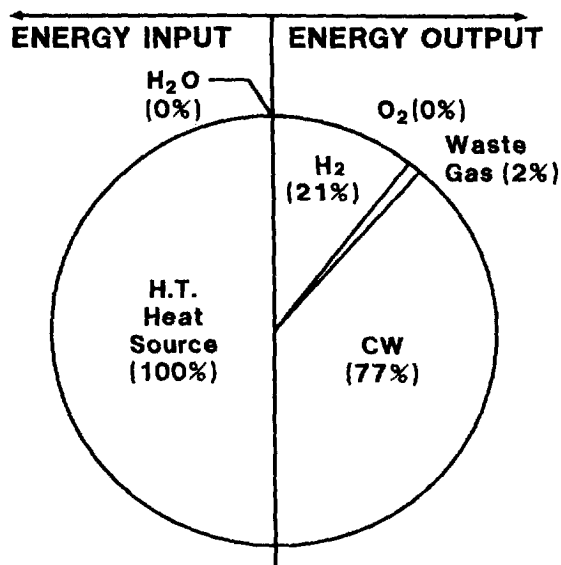


Fig. 2. (c) current-technology water electrolysis; (d) advanced-technology water electrolysis.

(e)



(f)

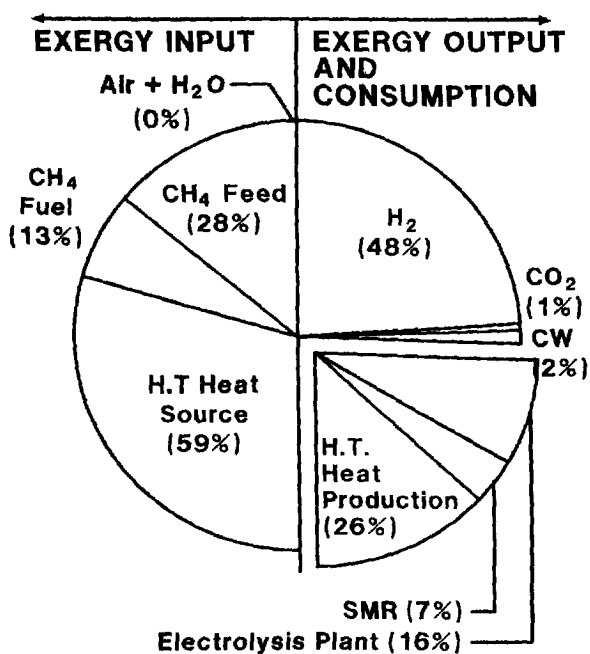
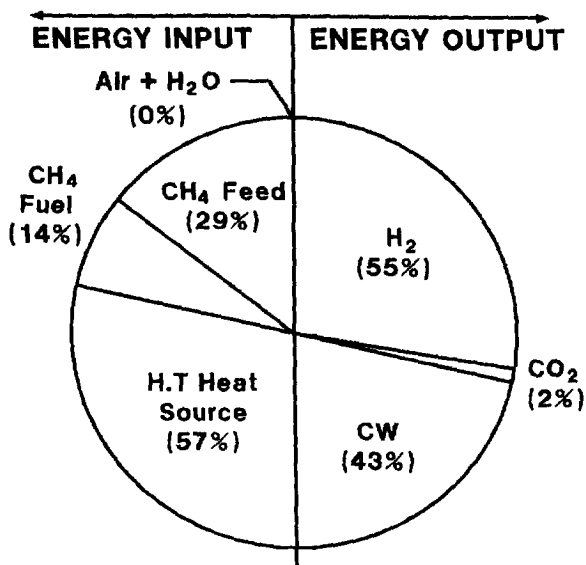
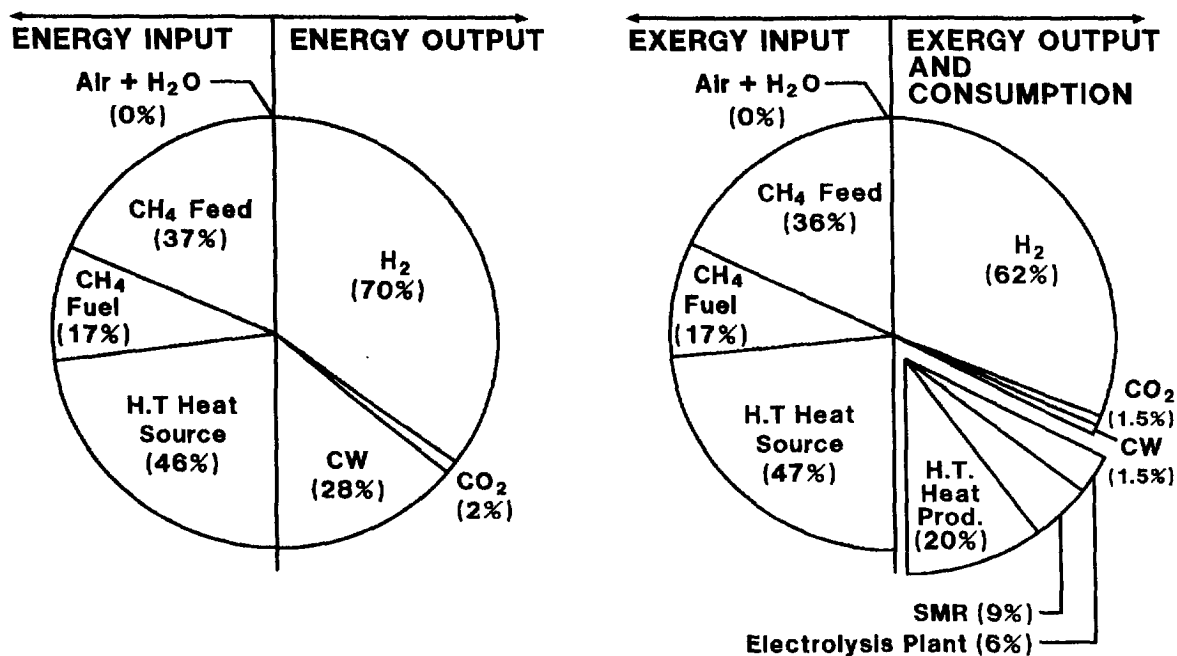


Fig. 2. (e) Thermochemical water decomposition; (f) SMR/current-technology water electrolysis.

(g)



(h)

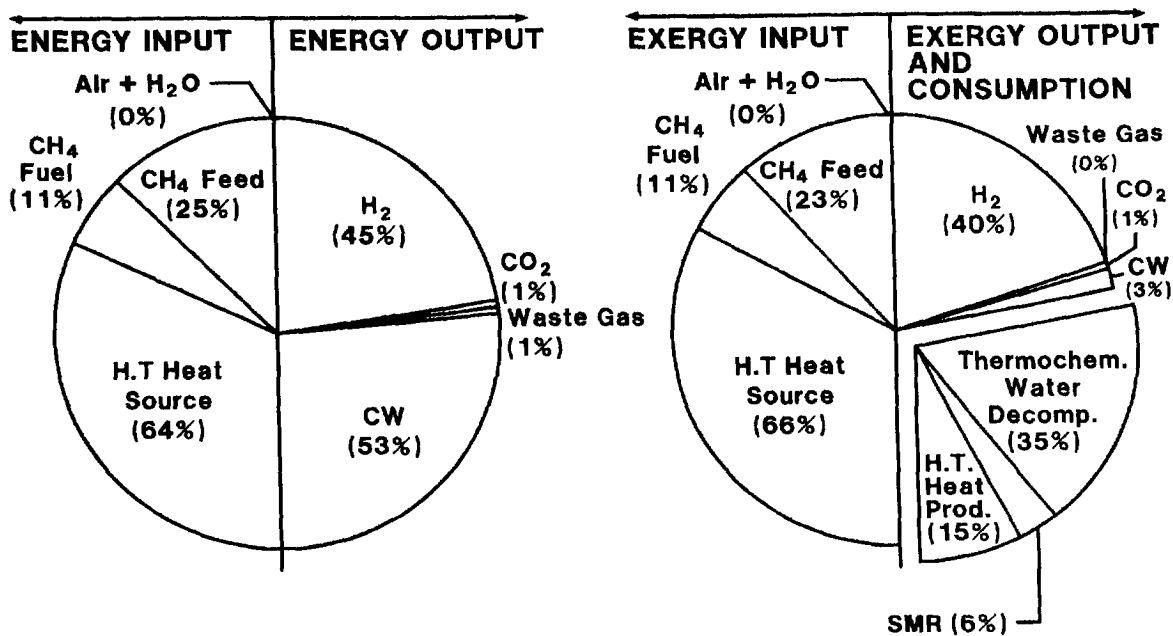


Fig. 2. (g) SMR/advanced-technology water electrolysis; (h) SMR/thermochemical water decomposition.

DISCUSSION

Hydrogen production from hydrocarbons

All the input energy and exergy to SMR and coal gasification are associated with the hydrocarbons (methane or coal). Most of the output energy is associated with waste cooling water, stack gas and hydrogen, and most of the output exergy with the hydrogen. For the hydrocarbon-based processes (and all other hydrogen production processes considered in this paper), little exergy is associated with the output cooling water because it is near the temperature of the environment, and the exergy associated with the stack gas is mainly chemical exergy.

The efficiencies (see Table 3) for these processes are relatively high, ranging (based on energy or exergy) approximately from 50% for coal gasification to 80% for SMR. Furthermore, since the hydrogen-to-carbon atomic ratios for methane (4) and coal (0.8) bracket the range covered by most of the hydrocarbons used in hydrogen production, the efficiencies for hydrogen production from hydrocarbons having intermediate ratios can be expected to lie between those determined here for SMR and coal gasification.

For each hydrocarbon-based process (and most of the other processes considered in this paper), the overall energy and exergy efficiencies are similar (see Table 3). However, these efficiencies differ for many plant devices. That is, the main energy and exergy losses are associated with different devices. In general, all energy loss is associated with waste emissions, while most (over 90% for all processes) of the exergy loss is associated with internal consumptions (see Table 4).

Cooling water and stack gas account for almost 100% of the energy losses and <10% of the exergy losses. Internal consumptions are responsible for most of the exergy losses. The main exergy consumptions for coal gasification occur in the gasifier (40% of the total exergy consumption) and combustor (32%), and for SMR in the combustor (50%). Another significant exergy consumption for both processes occurs in the CO₂-removal systems due to the use of steam to regenerate the CO₂ absorbent by heating. Remaining exergy consumptions are primarily attributable to compression, air separation and other chemical reactions and heat transfers.

The losses associated with air separation could be reduced using an integrated process where the oxygen requirement of the gasifier is met using by-product O₂ from water electrolysis or thermochemical water decomposition.

Hydrogen production from non-hydrocarbons

All of the input energy and exergy to current- and advanced-technology water electrolysis and thermochemical water decomposition is associated with high-temperature heat or the 'hypothetical heat source'. Most of the output energy is associated with waste cooling water and hydrogen, and most of the output exergy with the hydrogen.

The efficiencies (based on energy or exergy) range from low (ca 20%) for thermochemical water decomposition to high (over 90%) for water electrolysis. The water-electrolysis efficiencies for the advanced-technology case exceed those for the current-technology case. The exact values of the efficiencies for the processes depend on what sources of electricity and/or high-temperature heat are used.

Efficiencies for thermochemical water decomposition have been proposed in the literature which are higher than the ones calculated here for the Ispra Mark-10 process. These proposed efficiencies are often based on conceptual designs. The Ispra Mark-10 process was chosen because detailed data for a proposed commercial process existed.

When heat is considered as the primary input, cooling water accounts for nearly 100% of the energy losses, but only 3 to 7% of the exergy losses. The remaining exergy losses are associated with internal consumptions, mainly in:

- (i) the electrolysis unit and electrical generation plant for current-technology water electrolysis;
- (ii) the feedwater preheater and electrical generation plant for advanced-technology water electrolysis; and
- (iii) the primary reactors (where high-temperature heat is degraded to low-temperature heat) for thermochemical water decomposition.

Hydrogen production via integrated processes

The input energy and exergy to the integrated processes is associated with methane and high-temperature heat or the 'hypothetical heat source'. Most of the output energy is associated with waste cooling water and hydrogen, and most of the output exergy with the hydrogen.

The integrated processes reduce exergy losses for the independent water electrolysis and thermochemical water decomposition processes by using the oxygen emitted by them. Advantages of the integrated processes over the non-integrated ones include:

- (i) reduced requirements for methane fuel because the oxygen is not accompanied by nitrogen, which requires heating;
- (ii) increased overall efficiency;
- (iii) production of an additional by-product since, if the methane fuel is relatively pure, the stack gases are composed of relatively pure CO₂; and
- (iv) cleaner operation, since stack gas emissions are eliminated if the stack gas is contained as a by-product.

Each integrated-process efficiency is bracketed by the efficiencies for the separate processes comprising the integrated process. But each integrated-process efficiency is greater than the efficiency of the combined process in which the two separate processes are included but not integrated.

Energy losses are primarily associated with waste outputs, and exergy losses with internal consumptions.

When electricity and process heat are treated as the main inputs, exergy consumptions due to SMR are 46% of the total exergy consumptions for SMR/current-technology water electrolysis, 66% of the total for the SMR/advanced-technology water electrolysis and 15% of the total for the SMR/thermochemical water decomposition. When heat is treated as the main input, the above three values, respectively, decrease to 30%, 59% and 14%.

CONCLUSIONS

Several general trends are demonstrated. First, all energy losses in a process are associated with waste emissions (mainly with output cooling water), and most exergy losses with internal consumptions (mainly due to combustion and heat transfer across large temperature differences). Second, products of fuel production processes often have high energy and exergy contents, and wastes high energy and low exergy contents. Thus, energy analyses often indicate that wastes are valuable, and, to increase efficiency, quantities of waste effluents must be reduced, while exergy analyses indicate that wastes are not valuable and internal consumptions must be reduced to increase efficiency significantly. Third, overall efficiencies range widely (ca 20–90%) for the processes considered but, for any given process, the energy and exergy efficiency values are usually similar. This similarity occurs because input energy and exergy magnitudes are similar, and product energy and exergy magnitudes are similar. For other processes, where these conditions do not hold, values of energy and exergy efficiencies are different. Fourth, different devices are usually responsible for significant energy and exergy losses (e.g. typical efficiencies for boilers are 30% based on exergy and 85% based on energy).

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