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SYNFUEL (HYDROGEN) PRODUCTION FROM FUSION POWER

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ABSTRACT

A potential use of fusion energy for the production of synthetic fuel (hydrogen) is described. The hybrid-thermochemical bismuth-sulfate cycle is used as a vehicle to assess the technological and economic merits of this potential nonelectric application of fusion power.

POTENTIAL NONELECTRIC APPLICATIONS OF FUSION POWER have only recently been subjected to quantification by systems studies. Although fusion energy is furthest from practical and economic utilization compared to the other "inexhaustible" energy sources (fission, solar), studies and projections of magnetic, inertial and magneto-inertial fusion schemes for electric-power production have been performed at a significant level-of-effort since the early 1970's. Careful examination of energy resources, vectors, and end-uses projected for the mid-twentieth century, however, has led to preliminary consideration of nonelectric fusion applications. The energy end-use pattern that describes US consumption is depicted in Table I. Although the present 75% nonelectric consumption (now provided by hydrocarbon fuels) may diminish somewhat in the next century, this fraction is expected to remain high; for reasons of cost and/or technological feasibility, electricity simply cannot satisfy these nonelectric needs. If a serious mismatch between energy production capacities and utilization requirements is to be avoided, it seems prudent to consider nonelectric roles of fusion power, given that future scientific and technological progress is sufficient to permit utilization of fusion as a major energy source.

TABLE I. PRESENT PATTERN OF END-USE ENERGY CONSUMPTION IN THE UNITED STATES

| End Use | Percentage of Total |
|------------------------------------|---------------------|
| Electric Power | |
| Industrial | 14 |
| Residential and Commercial | 8 |
| Other | 2 |
| Total Electric | 24 |
| Fossil Fuels | |
| Transportation | 24 |
| Residential and Commercial Heating | 20 |
| Low-Temperature Process Heat | 17 |
| High-Temperature Process Heat | 9 |
| Conversion to Other Fuels | 7 |
| Nonenergy Uses | 5 |
| Other | 1 |
| Total Fossil Fuels | 76 |
| | 100 |

For this reason nonelectric or hybrid applications of fusion energy are considered an essential ingredient in the US fusion program.²

Examples of alternative fusion applications that have been subjected to preliminary examination include:

- high-temperature fixation of nitrogen in air to form ammonia.^{3,4}
- production of synthetic natural gas (SNG) and liquid fuels from coal.^{5,6}
- thermal splitting of carbon dioxide.⁷
- hydrogen production by water splitting using high-temperature electrolysis⁸ or thermochemical cycles.⁹
- supply of process heat for less specific applications.

Since a sustained thermonuclear reaction of deuterium and tritium (DT), to yield a 3.5-MeV alpha particle and a 14.1-MeV neutron, has not yet been controllably demonstrated to an extent required for an engineering energy breakeven, studies of nonelectric applications are generally based on highly conceptual reactor designs. Apart from the unique potential for producing large and potentially economic quantities of fissile fuel for use in a fission burner/convertor reactors^{10,11} a fusion energy source, as a producer of synthetic fuels (hydrogen, SNG, CO), chemical feedstocks (NH₃), or process heat, exhibits the following characteristics and possible advantages relative to other energy sources:

- 80% of the primary fusion energy is released as 14.1-MeV neutrons, which can be deposited as thermal energy in a relatively thin region (< 1 m) that is removed from the fusion source (i.e., a ~ 10-keV plasma).
- The primary fuel source (deuterium and lithium) is abundant.
- The DT plasma represents a strong source of electromagnetic radiation (~ 10-keV, ~ 1-Å wavelength) and charged particles (~ 0-10 keV DT ions, 0-3.5-MeV alpha particles), in addition to the 14.1-MeV neutrons.

Although direct radiolysis has been examined as a means to produce synthetic fuels,¹² most studies performed to date have considered only the generation of high-temperature heat in a blanket that surrounds the DT reaction chamber. Furthermore, because of neutron-induced radioactivity, direct nuclear heating of a chemical process stream, although offering significant thermodynamic and economic benefits, has yet to be seriously considered. Consequently, most studies have in common the design of high-temperature tritium-breeding (i.e., lithium containing) blankets that are heated primarily by the energetic fusion neutrons; the fusion energy is delivered to an

external chemical or electrochemical process by a high-temperature coolant or process-heat stream. Aside from the uniqueness of fuel supply, therefore, the major advantage perceived for fusion power that has received preliminary but quantitative examination is the potential to operate high-temperature blankets and therein generate process heat for use in an ex-reactor chemical process. This somewhat "conventional" approach also characterizes the studies which have so far examined the applicability of fusion power to the thermochemical or electrochemical splitting of water for generating hydrogen synfuel.

THERMOCHEMICAL HYDROGEN CYCLE

Hydrogen as a gaseous fuel supply can be manufactured using a variety of thermal energy sources, and, given the abundant resource (water), an inexhaustible and clean supply of thermal energy (breeder-fission, fusion-fission, solar, fusion) would result in a limitless and clean fuel supply. In addition to the well-known arguments made for a "hydrogen economy" (nonpolluting, transportable, easily stored, substitutable etc.), hydrogen is a valuable chemical commodity used in the production of ammonia and methanol and for ore reduction. For these reasons the applicability of fusion power to the generation of hydrogen has been examined.^{8,9} The production of hydrogen from water can be accomplished by direct thermal decomposition, electrolysis, thermochemical decomposition, or combinations thereof. Since the formation of water is characterized by a high enthalpy and free-energy of formation (-286 and -237 kJ/mol, respectively) a direct, one-step decomposition appears commercially impractical because of the unrealistically high temperature and low pressure required. The direct, ambient-temperature electrolysis of water occurs with an overall efficiency equal to the thermal-to-electric conversion efficiency reduced by the electrolyzer efficiency and, therefore, is ergonomically unattractive. Consequently, only the direct, but high-temperature, electrolysis (HTE) or the thermochemical decomposition (TCD) of water can potentially lead to increased thermal-conversion efficiencies. The potential for using fusion energy to drive a HTE process is being considered elsewhere,^{8,13} and the TCD approach is described here. The single most attractive feature of TCD is the reduction or elimination of electrical power generation and the potential for increased thermal efficiency (>50%). The HTE approach,^{8,13} on the other hand, would operate a direct electrolyser at elevated temperature and at a reduced free energy (and voltage), thereby achieving an increased thermal efficiency. For both HTE and TCD the interrelationship and trade-off between increased thermal efficiency and (increased) capital/operating costs remains an important and unresolved issue.

The status of TCD processes for hydrogen production has been reviewed,^{14,15} and Table II lists the TCD cycles that are under active research and/or development. In selecting a specific TCD or this study, the following practical issues were addressed:

Chemistry

- availability of accurate thermodynamic data
- availability of accurate kinetic data
- losses of intermediate compounds
- competing side reactions

Engineering

- matching of thermal energy source to the TCD cycle

TABLE II

THERMOCHEMICAL CYCLES UNDER ACTIVE RESEARCH AND DEVELOPMENT

CONTINUOUS-CIRCUIT, BENCH-SCALE TESTS:

- Hybrid Sulfuric Acid Cycle (Westinghouse Electric Corporation).
- Sulfuric Acid - Hydrogen Iodide Cycle (General Atomic Corporation).
- Hybrid Sulfuric Acid - Hydrogen Bromide Cycle (Euratom (Mark 13)).

ALTERNATE CYCLES UNDER RESEARCH:

- Bismuth Sulfate - Sulfuric Acid Cycle (Los Alamos Scientific Laboratory).
- Magnesium-Iodine Cycle (National Chemical Laboratory for Industry, Japan).
- Copper Sulfate Cycle (Institute of Gas Technology, USA).
- Potassium Iodide - Ammonia Cycle (Argonne National Laboratory).
- Barium Hydroxide - Copper Cycle (Oak Ridge National Laboratory).
- Zinc Selenide Cycle (Lawrence Livermore Laboratory).
- Calcium Bromide - Iron Oxide Cycles (University of Tokyo, Japan).

- development of high-temperature separation processes
- minimization of heat-exchange areas
- high-temperature, corrosive materials environment
- materials handling costs (capital and operations).

Available information on the few TCD cycles considered to date is inadequate to satisfy or to resolve these issues. Because of ongoing experimental support at LASL and the existence of a weak but developing and valuable data base,^{16,17} the bismuth-sulfate "hybrid" (i.e., a thermochemical cycle that operates with a low-temperature electrolysis step) TCD cycle was selected for study.⁹ Table III describes the attractions and problems associated with this cycle.

The use of $\text{SO}_2/\text{H}_2\text{SO}_4$ in the electrolyzer to reduce electrical power consumption by a factor of 2-3, in accordance with the first reaction depicted in Table III, appears in certain hybrid TCD cycles. Sulfuric acid is also generated, although by a different reaction, in "pure" thermochemical cycles such as the sulfuric-acid/iodine cycle under development at the General Atomic Corporation (Table II). In order to resolve problems of SO_2 recovery (i.e., H_2SO_4 handling, evaporation and decomposition), a metal (bismuth) oxide or sub-sulfate would be added to the acid anolyte, and a metal sulfate would be precipitated from the acid solution. The bismuth sulfates so formed would be dried and thermally decomposed by the fusion-generated energy, according to the (step 3) reaction sequence depicted on Table III. In addition to possessing a desirable chemical reactivity, (i.e., short reaction times and small chemical reactors) bismuth sulfate was selected because the absence of hydrates, which would lead to an undeniably and difficult drying step. The SO_3 evolved from the sulfate decomposition steps would be thermally reduced to SO_2 using the thermal power from the fusion driver; the $\text{SO}_3/\text{SO}_2/\text{O}_2$ mixture would, in fact, serve as the primary heat-exchange medium that powers the bismuth-sulfate decomposers.⁹ The high acid concentration (52.7 wt%) required to regenerate the

TABLE III
HYBRID BISMUTH SULFATE THERMOCHEMICAL CYCLE

● Three Step Cycle:

| | |
|--|----------------------|
| 1) $\text{SO}_2(\text{g}) + 2\text{H}_2\text{O}(\text{l}) \rightarrow \text{H}_2\text{SO}_4(\text{sol}) + \text{H}_2(\text{g})$ | 350 K (Electrolysis) |
| 2) $\text{H}_2\text{SO}_4(\text{sol}) + 1/3 \text{Bi}_2\text{O}_3(\text{s}) \rightarrow 1/3 \text{Bi}_2\text{O}_3 \cdot 3\text{SO}_3 + \text{H}_2\text{O}(\text{l})$ | 350 K |
| 3) $1/3 \text{Bi}_2\text{O}_3 \cdot 3\text{SO}_3(\text{s}) \rightarrow 1/3 \text{Bi}_2\text{O}_3(\text{s}) + \text{SO}_2(\text{g}) + 1/2 \text{O}_2(\text{g})$ | 900-1250 K |

● Data on Step 3

| | T(K) | $\Delta H(\text{kJ/mol})$ | Time(m) |
|---|----------|---------------------------|---------|
| $\text{Bi}_2\text{O}_3 \cdot 3\text{SO}_3(\text{s}) \rightarrow \text{Bi}_2\text{O}_3 \cdot 2\text{SO}_3(\text{s}) + \text{SO}_3(\text{g})$ | 876 | 161 | 1.5 |
| $\text{Bi}_2\text{O}_3 \cdot 2\text{SO}_3(\text{s}) \rightarrow \text{Bi}_2\text{O}_3 \cdot \text{SO}_3(\text{s}) + \text{SO}_3(\text{g})$ | 973(?) | 167(?) | 3.0 |
| $\text{Bi}_2\text{O}_3 \cdot \text{SO}_3(\text{s}) \rightarrow \text{Bi}_2\text{O}_3 \cdot 1/3\text{SO}_3(\text{s}) + 1/3\text{SO}_3(\text{g})$ | 1223 | ? | 10(+) |
| $\text{Bi}_2\text{O}_3 \cdot 2/3\text{SO}_3(\text{s}, \text{l}) \rightarrow \text{Bi}_2\text{O}_3(\text{l}) + 2/3\text{SO}_3(\text{g})$ | 1253 | ? | ? |
| $\text{SO}_3(\text{g}) \rightarrow \text{SO}_2 + 1/2\text{O}_2(\text{g})$ | 960-1500 | 96.3 | rapid |

● Attractions

- Replace acid drying with (insoluble) metal sulfate precipitation
- H_2 produced in a pure state
- Metal sulfate does not form hydrates (e.g., Cu and Al form hydrates)
- Acid handling occurs at low-to-moderate temperatures
- Electrolysis occurs at low acid concentrations (lower voltage, higher efficiency)
- Possible use of chemical heat-pipe effect for $\text{SO}_3 \rightarrow \text{SO}_2 + 1/2\text{O}_2$ reaction

● Crucial Issues:

- Large volume of solids handling
- Decomposition chemistry not fully resolved
- Reaction rates and heat transfer in $\text{Bi}_2\text{O}_3 \cdot x\text{SO}_3$ decomposers must be rapid to minimize size
- Recuperation of latent heat from drying of solids
- Low-voltage electrolyzer must be demonstrated under production conditions

sulfate represents a disadvantage in that high electrolytic voltages (> 0.6 V) result. Furthermore, if the $\text{Bi}_2\text{O}_3 \cdot 3\text{SO}_3$ is reduced completely to the oxide (melting point - 977 K), a molten oxide and a complicated handling problem is incurred. If the sulfate reduction is allowed to proceed only to the $\text{Bi}_2\text{O}_3 \cdot 2/3\text{SO}_3$ (Table III), a liquid phase does not form below 1253 K. In addition, the endothermic energy required to remove one mol of SO_3 from the sulfate was assumed to equal to one-third of the measured full-decomposition energy⁹ (250 kJ/mol H_2); recent experimental evidence suggests the removal of the first two mols of SO_3 from $\text{Bi}_2\text{O}_3 \cdot 3\text{SO}_3$ proceeds with a reduced heat of reaction (167 kJ/mol per SO_3), and operation with a partial sulfate decomposition would result in increased cycle efficiency because of the reduced endothermal heat requirement. Furthermore, a reduced acid concentration in principle may lead to decreased electrolytic voltages and increased thermal efficiency¹⁸; experimental electrochemical data for the lower acid concentrations are not yet available. Generally, operation at lower acid concentrations and lower sulfates leads to significant benefits from both the viewpoint of cycle efficiency and decomposer design compared to an earlier design.⁹ These benefits are derived at the cost of an increased solids handling requirement per unit of hydrogen production. It is noted on Table III that the reaction times for all decomposition steps of interest are favorable from the viewpoint of process equipment sizes.

THERMOCHEMICAL PROCESS DESIGNS

A preliminary process design based on the full decomposition sequence (Table III) and an electrolysis at 52.7 wt% H_2SO_4 has been reported.⁹ The process modifications described in the previous section have been incorporated into a newer design that promises higher thermal efficiencies and reduced materials problems. Figure 1 and Table IV show the results of a parametric study of the influence of the peak operating temperature T(R), electrolytic cell voltage $E_{\text{CELL}}(\text{V})$, and endothermic heat requirement Q(kJ/mol H_2) on the earlier reference design efficiency.⁹ Although only small

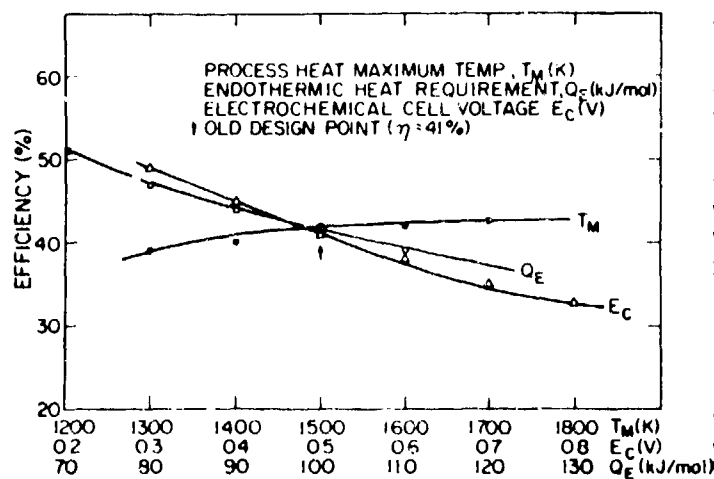


Fig. 1. Parametric dependence of thermochemical hydrogen cycle efficiency on maximum operating temperature, electrolytic cell voltage, and endothermic heat requirement.

increases in cycle efficiency result from increased process temperatures, significant improvements would result if the electrolytic cell voltage and/or the endothermic heat requirement could be reduced.

A preliminary energy balance incorporating these features is the basis of a new design¹⁹ in which two major modifications to the process result in a significant efficiency increase (Table IV). The endothermic energy requirement (per mole of H_2) for both the bismuth sulfate decomposition step and the sulfur trioxide decomposition totals 251 kJ/mol, compared to the earlier value of 379 kJ/mol. This decrease is accomplished by the removal of 1 to 4/3 mols of SO_3 per mol of H_2 rather than three mols of SO_3 . This benefit is derived, however, at the cost of slightly increased equipment size and

Table IV. PROCESS ENERGY BALANCE FOR HYBRID BISMUTH SULFATE THERMOCHEMICAL CYCLE

| Process Steps | Heat Required kJ/mol H ₂ | Heat Available kJ/mol H ₂ | Work Heat Equiv. kJ/mol H ₂ |
|--|--|---|---|
| Decomposer | | | |
| Bi ₂ O ₃ ·2SO ₃ + Bi ₂ O ₃ ·SO ₃ + SO ₃ | 167 | | |
| | 875-1250K | | |
| SO ₃ + SO ₂ + 1/2O ₂ | 95 | | |
| | 1500K | | |
| Bi ₂ O ₃ ·2SO ₃ preheat | 243 ^(a) | | |
| | 350-1250K | | |
| SO ₃ preheat | 19 | | |
| | 1250-1500K | | |
| Bi ₂ O ₃ ·SO ₃ cooling | | 179 ^(a) | |
| | | 1250-350K | |
| SO ₂ cooling | | 75 | |
| | | 1500-350K | |
| 1/2O ₂ cooling | | 19 | |
| | | 1500-350K | |
| TOTALS | 524 | 273 | |
| NET (Q ₁) | 251 | | |
| Electrolyzer | | | |
| 2H ₂ O + SO ₂ + H ₂ SO ₄ + H ₂ | | | 228 ^(b) |
| (0.45 V, 2000 A/m ² , 3 wt% H ₂ SO ₄) | | | |
| TOTALS (Q ₂) | 228 | | |
| Bismuth Sulfate Reaction | | | |
| Bi ₂ O ₃ ·SO ₃ + H ₂ SO ₄ | | (75) | |
| Bi ₂ O ₃ ·2SO ₃ + H ₂ O | | 350K | |
| Bi ₂ O ₃ ·2SO ₃ drying | | | |
| 5 H ₂ O preheat | 61 | | |
| | 350-510K | | |
| 5 H ₂ O vaporization | 162 | | |
| | 510K | | |
| 5 H ₂ O condensation | | 172 | |
| | | 485K | |
| TOTALS | 223 | 172 | |
| NET (Q ₃) | 51 | | |
| SO₂/O₂ Separator | | | |
| SO ₂ compression | | | 42 ^(b) |
| O ₂ expansion | | 10 ^(b) | |
| SO ₂ absorbers | 14 | | |
| | 600-700K | | |
| TOTALS | 14 | 10 | 42 |
| NET (Q ₄) | 46 | | |

(a) estimated

(b) To obtain the heat equivalent of work, the work term is divided by 0.38, which is the assumed conversion efficiency of converting 800 K heat to work.

$$\text{Net Heat Requirement} = \sum Q_i = Q_1 + Q_2 + Q_3 + Q_4 = 251 + 228 + 51 + 46 = 576 \text{ kJ/mol H}_2$$

$$\text{Cycle thermal efficiency, } \eta = 286/576 = 0.50$$

capital/operating cost. The electrolytic voltage has been taken as 0.45 V. This voltage results in a thermal energy requirement of 228 kJ/mol H₂, compared to the previous value of 253 kJ/mol. In principle, electrolysis at a reduced acid concentration should require less energy than electrolysis at a much higher acid concentration of 52.7 wt%.

Overvoltage associated with cell resistance should be partly overcome in addition to a reduction in theoretical voltage, resulting in an overall voltage decrease. The total thermal energy requirement for the modified LASL bismuth sulfate cycle presently equals 576 kJ/mol H₂, resulting in a process thermal efficiency of 50%. This thermal efficiency is supported by detailed process calculations based on an engineering flowsheet for the LASL thermochemical cycle.¹⁹

Figure 2 depicts schematically the chemical and energy flow envisaged for this fusion-driven TCD cycle. As noted previously, sensible heat from the fusion driver would be deposited directly into the SO₃/SO₂/O₂ stream, which in turn would supply high-temperature process heat to the sulfate decomposition. This approach differs considerably from the earlier design,⁹ in which the fusion energy was delivered to the SO₃/SO₂/O₂ stream by means of a primary helium coolant (1500/800 K, 1 MPa) and heat exchanger. The use of a helium-cooled graphite outer blanket (1500 K) in conjunction with a lower temperature (800 K) lithium-cooled inner blanket for the fusion power plant lead to a poor energy match between the electrical requirements (fusion-driver recirculating power, electrolyzer power and compressors used in the chemical process) and the high-temperature process heat demanded by the thermochemical cycle. For this reason, a conceptual "lithium boiler" fusion blanket²⁰ has been adopted in order to increase the proportion of fusion energy delivered as high-temperature process heat, to eliminate the need for a secondary heat exchanger, and to breed tritium in a high-temperature containment. As indicated in Fig. 2, conversion of the lower-temperature (first-wall) thermal energy to electricity, as well as direct-conversion energy (a tandem mirror fusion reactor is being considered),²¹ supplies all electrical requirements of both the thermochemical and the fusion power plants.

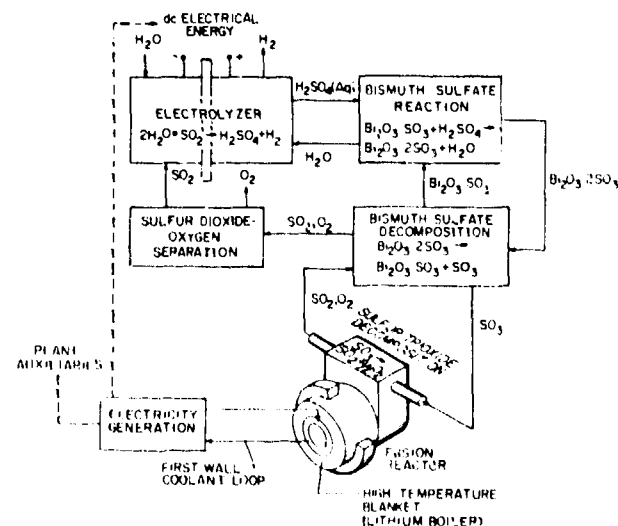


Fig. 2. Schematic diagram of bismuth-sulfate/sulfuric-acid thermochemical hydrogen cycle driven by a fusion reactor. Refer to Table III for details of reaction sequence.

ECONOMIC CONSIDERATIONS

The design for both the fusion driver and the thermochemical plant is not sufficiently resolved to permit a detailed cost estimate. Consequently, estimates of hydrogen production costs have been limited to simple parametric analyses²² in order to identify major cost drivers. Generally, above a minimum hydrogen selling price and for the same unit investment and thermochemical cycle efficiency, lower production costs result when the fusion energy utilized to satisfy purely thermal requirements can be maximized. The hydrogen production costs vary hyperbolically with cycle efficiency and linearly with the cost of the fusion driver. Comparisons on an absolute basis have been made with recent results for other nuclear hydrogen production costs.²³ These studies indicate comparable values if the fusion driver and thermochemical plant cost, respectively, cost no more than 1-2 and 0.6-1 times the present cost of a nuclear fission plant, if the thermochemical cycle efficiency is maintained above 50%. Based on this simple parametric analysis, typical hydrogen production costs would fall in the range 7-13 \$/GJ, which is still a factor of 3-5 above present methane costs but is comparable to the cost of other proposed sources of nuclear hydrogen.

CONCLUSIONS

- The major advantage that fusion energy offers as a producer of synfuel, aside from the abundance of the fusion fuel-resource, is the potential for generation of process heat at temperatures above those presently thought feasible in a fission blanket.
- Most studies of nonelectric applications of fusion energy, excluding fissile-fuel production, have emphasized in one form or another the generation of high-temperature process heat.
- Use of a partial decomposition of $\text{Bi}_2\text{O}_3 \cdot 2\text{SO}_3$ and a lowering of the H_2SO_4 acid concentration leads to an increase of thermochemical cycle efficiency at the expense of increased mass-handling requirements per unit of hydrogen produced.
- The generation of tritium in a low-temperature lithium blanket adversely affects the thermal performance of the global system, leads to a poor thermal energy match, and generally forces the system to generate excess electricity.
- Materials problems within both the fusion driver and the chemical process are endemic to most alternative applications which specify temperatures in excess of 1400-1500 K to achieve economically attractive and competitive efficiencies.
- Although parametric and unsubstantiated by detailed design, estimates of hydrogen production costs indicate that the fusion-driven thermochemical hydrogen system are competitive with other approaches to synthetic hydrogen. These production costs, however, are a factor of 3-5 above present methane costs.

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