Hydrogen Production Using Electrolysis Versus a Hybrid Cycle

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Economic Comparison of Hydrogen Production Using Sulfuric Acid Electrolysis and Sulfur Cycle Water Decomposition

EM-789
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Final Report, June 1978

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ABSTRACT

An evaluation of the relative economics of hydrogen production using two advanced techniques was performed. The hydrogen production systems considered were the Westinghouse Sulfur Cycle Water Decomposition System and a water electrolysis system employing a sulfuric acid electrolyte. The former is a hybrid system in which hydrogen is produced in an electrolyzer which uses sulfur dioxide to depolarize the anode. The overall reaction in this cell is:

The electrolyte is sulfuric acid. Development and demonstration efforts have shown that extremely low cell voltages can be achieved; recent test runs have shown total cell voltages of 600 mV for a cell operating at 200 mA/cm^2 and 50°C at atmospheric pressure. The second system uses a similar sulfuric acid electrolyte technology in water electrolysis cells, i.e., cells producing both hydrogen and oxygen.

The comparative technoeconomics of hydrogen produced by the hybrid Sulfur Cycle and by water electrolysis using a sulfuric acid electrolyte were determined by assessing the performance and economics of 380 million SCFD plants, each energized by a very high temperature nuclear reactor (VHTR). The VHTR, having a helium coolant outlet temperature of 1850°F , is configured to provide the thermal and/or electrical needs of the hydrogen production plants.

The evaluation concluded that the overall efficiencies of hydrogen production, for operating parameters that appear reasonable for both systems, are approximately 41 percent for the sulfuric acid electrolysis and 47 percent for the hybrid Sulfur Cycle. The economic evaluation of hydrogen production, based on a 1976 cost basis and assuming a developed technology for both hydrogen production systems and the VHTRs, indicated that the hybrid Sulfur Cycle could

generate hydrogen for a total cost approximately six to seven percent less than the cost from the sulfuric acid electrolysis plant. This difference in cost may not be significant in light of the uncertainties that surround many of the technoeconomic assessment. The difference in efficiency, because of its magnitude, is more likely to represent a real difference despite the uncertainties.

EPRI PERSPECTIVE

PROJECT DESCRIPTION

This final report discusses a technoeconomic comparison of the Westinghouse hybrid cycle with a water electrolysis process wherein the same electrolyzer concept was used for both processes and, in as far as possible, similar assumptions regarding energy source, fuel cost, etc. were employed in both cases. The objective was to better establish the economics of the two approaches and to establish the hydrogen production technology areas for R&D support.

PROJECT OBJECTIVES

The near-term objective of the EPRI hydrogen program is the identification of hydrogen applications and production technologies that can benefit the electric utility industry in the future. Under RP320 and RP467, EPRI assessed the technoeconomics of producing hydrogen by water electrolysis (using off-peak power) and thermochemical water-splitting cycles, respectively. Those results had indicated that water electrolysis would result in lower cost hydrogen than the pure thermochemical cycles assessed at that time. An alternative concept is a hybrid (thermochemical cycle with an electrolytic step) cycle being developed by Westinghouse.

CONCLUSIONS AND RECOMMENDATIONS

This report concludes that although the Westinghouse hybrid cycle has a better overall efficiency than water electrolysis (47% vs. 41%), the costs of the hydrogen are essentially the same at \$7.30 - \$7.80/10⁶ Btu (1976 dollars). Considering the major uncertainties that advanced thermochemical cycles face, such as the speculative nature of the high temperature heat source, as well as the kinetics of key reactions, EPRI does not recommend a large involvement in the thermochemical process area at this time.

It is possible that the General Electric solid polymer electrolyte (SPE) electrolyzer technology could be applied to the Westinghouse hybrid cycle with an improvement in both efficiency and cost. EPRI is now considering a project that would assess that possibility.

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Section 1

EXECUTIVE SUMMARY

The production of hydrogen from non-fossil energy sources for use as a fuel, chemical reagent, or energy storage and transport medium has been gaining increased attention over the past several years. This interest reflects not only the concern with the availability and cost of the fossil resources, e.g., natural gas, on which today's bulk hydrogen production is based, but also reflects the desire to maximize the efficiency of energy resource utilization and minimize the environmental impact of power production and utilization. Hydrogen production can provide a mechanism whereby our more plentiful resources, such as nuclear or solar power, can be converted in an environmentally acceptable manner into a transportable and storable energy form. The hydrogen can ultimately be converted into electric power by, for example, fuel cells, or used as a feedstock in processes as diverse as fertilizer manufacture to synthetic hydrocarbon fuel production. The potential of hydrogen utilization in these manners has led to research, development, and demonstration programs being undertaken to investigate advanced electrochemical, thermochemical, and hybrid electrochemical/thermochemical hydrogen production processes.

Westinghouse has been involved in the development of a hybrid electrochemical/thermochemical hydrogen production process called the Sulfur Cycle Water Decomposition System. The work conducted by them (1, 2) and others (3) suggests that this hybrid process may be more economical than alternative thermochemical hydrogen production systems. Electrochemical hydrogen production systems, on the other hand, are receiving considerable attention for their capability to produce hydrogen in a less complex process.

A key constituent of the Westinghouse Sulfur Cycle hybrid system is a step in which hydrogen is produced in an electrolyzer which uses sulfur dioxide to depolarize the anode. The overall reaction in this cell is:

The electrolyte in the cell is sulfuric acid. Development and demonstration efforts have shown that extremely low cell voltages can be achieved; recent test runs have shown total cell voltages of 600 mV for a cell operating at 200 mA/cm² and 50°C at atmospheric pressure. A similar sulfuric acid electrolyte concept is also applicable to water electrolysis cells, i.e., cells producing both hydrogen and oxygen.

It is the purpose of this study to determine the comparative technoeconomics of hydrogen produced both by the hybrid Sulfur Cycle and by water electrolysis using a sulfuric acid electrolyte.

SULFUR CYCLE HYDROGEN PRODUCTION PLANT

The Sulfur Cycle Hydrogen Production Plant is based on a conceptual design and cost estimate prepared for NASA (1) and updated for the U. S. Department of Energy (2). The plant is a grass roots, self-sufficient 380 million SCFD hydrogen production unit energized by a very high temperature nuclear reactor (VHTR).

In the Sulfur Cycle, hydrogen and sulfuric acid are produced electrolytically by the reaction of sulfur dioxide and water. The process is completed by vaporizing the sulfuric acid and thermally reducing, at higher temperatures, the resultant sulfur trioxide to sulfur dioxide and oxygen. Following separation, sulfur dioxide is recycled to the electrolyzer and oxygen is either utilized or vented.

As in conventional water electrolysis, hydrogen is produced at the electrolyzer cathode. Unlike conventional electrolysis, sulfuric acid, rather than oxygen, is produced at the anode. Operation in this manner substantially reduces the theoretical power required per unit of hydrogen production over that required in water electrolysis. This is partially offset, however, by the need to add thermal energy to the process in the acid vaporizer and the sulfur trioxide reduction reactor.

The overall process flowsheet for the water decomposition system is shown in Figure 1-1. The energy source for the water decomposition system is a very high temperature nuclear reactor (VHTR) producing both electric power and a high temperature helium stream to the process. Within Battery G, electrical power, water, and recycled sulfur dioxide are consumed to produce hydrogen and sulfuric acid. The hydrogen, of electrolytic purity, is withdrawn as product while the sulfuric acid is sent to Battery H. Using thermal energy from the VHTR, the acid in Battery H is vaporized to produce a mixture of steam and sulfur trioxide. The sulfur trioxide is thermal-catalytically reduced at higher temperatures to produce sulfur dioxide and oxygen. These gases are separated within Battery I. The sulfur dioxide is recycled to Battery G and the oxygen is available as a by-product for sale. The only major consumable for the process is water. Small quantities of make-up are required, for example, to compensate for leakage and losses, catalyst deactivation, etc. The sulfur oxides are recycled and all process and plant energy needs are provided by the VHTR.

The performance level of the SO_2 depolarized electrolyzer is, of course, key to the efficiency and economics of hydrogen production by the Sulfur Cycle. The development activities underway will determine the ultimate capability of the system; for the purpose of this study the electrolyzer performance; i.e., total cell voltage, was permitted to vary parametrically between 400 and 800 mV. The electrolyzer parameters selected for the evaluation are shown in Table 1-1.

Table 1-1
SULFUR CYCLE ELECTROLYZER PARAMETERS

Type	SO_2 Depolarized Anode
Electrolyte	Sulfuric Acid
Acid Concentration	80%
Temperature	100°C
Pressure	20 ATM
Current Density	200 mA/cm ²
Cell Voltage	400 - 800 mV

Figure 1-1. Overall Process Schematic of the Sulfur Cycle Decomposition System

The very high temperature nuclear reactor (VHTR), suitable for use with the water decomposition system, is predicated on the integration of the technologies from the NERVA nuclear rocket engine program and land-based gas cooled reactor programs into an advanced graphite moderated, helium cooled reactor (4). The VHTR plant produces both very high temperature (1850°F) heat and electrical power for the hydrogen production plant. The heat is transported to the process via an intermediate heat transfer loop at temperatures sufficiently high to permit a peak process temperature of 1600°F.

The performance analysis of the Sulfur Cycle plant shows that the overall efficiency of hydrogen production is approximately 47 percent when the electrolyzer cell is operating at 600 mV. The variation in efficiency over the cell voltages considered in the study is shown in Figure 1-2.

The estimated cost of the plant, in 1976 dollars, is about \$1.22 billion for the situation where a 600 mV cell voltage is attained. Figure 1-3 shows the variation in total plant investment as a function of the cell voltage, as well as the proportions of the total cost contributed by the VHTR and hydrogen production parts of the plant.

The hydrogen "gate price", representing the annual charges on invested capital, operation and maintenance costs, and nuclear fuel costs are shown in Figure 1-4 for an assumed 80 percent capacity factor. Again, for a reasonably expected 600 mV cell voltage and a nuclear fuel cost of \$0.50 per million Btu, hydrogen would be produced at a cost of \$2.38 per thousand standard cubic feet, or \$7.31 per million Btu.

SULFURIC ACID ELECTROLYSIS PLANT

Process flow sheets, mass and energy balances, and cost estimates were prepared for an identically sized grass roots self-sufficient hydrogen production plant using water electrolysis with a sulfuric acid electrolyte. The electrolysis cell configuration, current densities, and cost assumptions were, wherever possible, identical to those for the Sulfur Cycle electrolyzer to make the technoeconomic comparison as consistent as possible. Table 1-2 shows the major technical parameters selected for the electrolysis cell.

Figure 1-2. Sulfur Cycle Overall Plant Efficiency Versus Cell Voltage

Figure 1-3. Sulfur Cycle Plant Capital Cost Variation with Cell Voltage - 1976 Basis

Figure 1-4. Sulfur Cycle Plant Hydrogen "Gate Price"
Versus Nuclear Fuel Cost - 1976 Basis

Table 1-2
SULFURIC ACID WATER ELECTROLYZER

ELECTROLYTE	SULFURIC ACID
ACID CONCENTRATION	28%
TEMPERATURE	100°C
PRESSURE	20 ATM
CURRENT DENSITY	200 mA/cm ²
CELL VOLTAGE	1.58-1.78 V

As in the Sulfur Cycle analysis, the cell voltage was allowed to vary parametrically over a range that is believed representative of the possible operating regime of a developed cell.

The hydrogen plant flow sheet, as shown in Figure 1-5, is typical of water electrolysis plants, except that one is handling a sulfuric acid, rather than an alkaline, electrolyte. The power source for the hydrogen plant is a very high temperature nuclear reactor (VHTR), with the same technological level as the plant for the Sulfur Cycle, but using the 1850°F helium coolant outlet stream in a high efficiency Brayton cycle. At these temperature levels, and with the gas turbine cycle selected, a net power generation efficiency of 46.1 percent can be achieved. The overall hydrogen plant efficiency combines the efficiency of electric power generation with the efficiency of the electrolysis plant, and is very much a function of cell voltage. For an assumed cell operating voltage of 1.68 volts, the electrolysis plant efficiency would be about 88.6 percent and the overall hydrogen plant efficiency about 40.9 percent. Figure 1-6 shows how this efficiency varies with cell voltage.

The total investment in the plant, including both the VHTR electric generating unit and the electrolysis plant, is shown in Figure 1-7 as a function of cell voltage. For a plant operating at 1.68 volts, the total capital investment is about \$1.28 billion. The cost make-up of this plant, although in total not too different than the Sulfur Cycle plant, shows the hydrogen production plant to be much smaller and the nuclear energy source much larger than the corresponding items in the Sulfur Cycle Plant.

Figure 1-5. H_2 Production Plant Schematic - Sulfuric Acid Electrolysis

Figure 1-6. Sulfuric Acid Electrolyzer Overall Plant Efficiency Versus Cell Voltage (100°C)

Figure 1-7. Sulfuric Acid Electrolyzer Plant Capital Cost Versus Cell Voltage - 1976 Basis (100°C)

The production cost of the hydrogen, based on an 80 percent plant capacity factor, is shown in Figure 1-8 as a function of both fuel cost and cell voltage. For an assumed 1.68 volt operating condition and a \$.50 per million Btu nuclear fuel cost, the hydrogen "gate price" would be \$2.54 per thousand standard cubic feet or \$7.80 per million Btu.

COMPARATIVE TECHNOECONOMICS

In comparing the performance and economics of the two hydrogen production systems, it must be kept in mind that both systems, and the nuclear energy source for both, require further development to substantiate the engineering judgments used in defining flow sheets, performance capabilities, component designs, and economics.

The results of the evaluation of the Sulfur Cycle and sulfuric acid electrolysis plant show a consistent difference in overall thermal efficiency for the two concepts, with the Sulfur Cycle having an efficiency some six percentage points higher (47 percent vs 40.9 percent for reasonably expected operating conditions) than the sulfuric acid electrolysis plant. Figure 1-9 shows the comparative efficiencies of the two systems as a function of attainable cell voltage.

The relative "gate price" of hydrogen from the two systems, as a function of fuel cost, is shown in Figure 1-10. The Sulfur Cycle is shown to have a production cost approximately six to seven percent less than the sulfuric acid electrolysis plant. Because of the lower efficiency of the latter, the effect of increasing fuel costs is to also increase the production cost difference between the two systems.

Finally, the performance level at which the two systems become cost competitive can be determined from Figure 1-11. It can be shown, for instance, that if the Sulfur Cycle electrolyzer were to operate at 600 mV, the sulfuric acid electrolysis plant would have to have a cell voltage of about 1.43 volts to be competitive. Since this is below the thermoneutral voltage, it is probably not a practical operating point.

Figure 1-8. Sulfuric Acid - Electrolyzer Plant Hydrogen
"Gate Price" Versus Nuclear Fuel Cost - 1976 Basis

Figure 1-9. Comparison of Overall Plant Efficiencies Versus Cell Voltage

Figure 1-10. Sulfur Cycle Plant Comparison with Sulfuric Acid Electrolyzer Plant Hydrogen "Gate Price" Versus Nuclear Fuel Cost

Figure 1-11. Comparison of Hydrogen "Gate Price" Versus Cell Voltage - 1976 Basis (\$0.50/10⁶ Btu Nuclear Fuel Cost)

CONCLUSIONS

The results of this study, using a consistent design and costing approach for both systems, show that:

- The overall efficiency of hydrogen production by the hybrid Sulfur Cycle Water Decomposition System is higher than that which can be achieved in a plant using water electrolysis based upon a sulfuric acid electrolyte.
- The economics, recognizing the caveats of cost estimates on systems and components not yet fully developed, show marginally lower hydrogen production costs for the Sulfur Cycle hydrogen production plant.
- The development requirements for the nuclear energy source are essentially the same regardless of which hydrogen production system is employed.
- The electrolysis step is a critical element of both hydrogen production systems. The cost and efficiency of the electrolysis step is, in both systems, a major determinant of hydrogen cost and overall system efficiency.
- The development programs required to commercialize the hydrogen production systems themselves are substantially different. The sulfuric acid electrolysis system will require less funding and time to achieve its objectives, particularly since it does not have to contend with the high temperature process steps and materials for high temperature applications required in the Sulfur Cycle. While not considered during this study, a sulfuric acid electrolysis system could, of course, be driven by existing methods of power generation that require no development.

Section 2

INTRODUCTION

The production of hydrogen from non-fossil energy sources for use as a fuel, chemical reagent, or energy storage and transport medium has been gaining increased attention over the past several years. This interest reflects not only the concern with the availability and cost of the fossil resources, e.g., natural gas, on which today's bulk hydrogen production is based, but also reflects the desire to maximize the efficiency of energy resource utilization and minimize the environmental impact of power production and utilization. Hydrogen production can provide a mechanism whereby our more plentiful resources, such as nuclear or solar power, can be converted in an environmentally acceptable manner into a transportable and storable energy form. The hydrogen can ultimately be converted into electric power by, for example, fuel cells, or used as a feedstock in processes as diverse as fertilizer manufacture to synthetic hydrocarbon fuel production. The potential of hydrogen utilization in this manner has led to research, development, and demonstration programs being undertaken to investigate advanced electrochemical, thermochemical, and hybrid electrochemical/thermochemical hydrogen production processes.

Westinghouse has been involved in the development of a hybrid electrochemical/thermochemical hydrogen production process called the Sulfur Cycle Water Decomposition System. The work conducted by them (1,2) and others (3) suggests that this hybrid process may be more economical than alternative thermochemical hydrogen production systems. Electrochemical hydrogen production systems, on the other hand, are receiving considerable attention for their capability to produce hydrogen in a less complex process.

A key constituent of the Westinghouse Sulfur Cycle hybrid system is a step in which hydrogen is produced in an electrolyzer which uses sulfur dioxide to depolarize the anode. The overall reaction in this cell is

The electrolyte in the cell is sulfuric acid. Development and demonstration efforts have shown that extremely low cell voltages can be achieved; recent test runs have shown total cell voltages of 600 mV for a cell operating at 200 mA/cm² and 50°C at atmospheric pressure. A similar sulfuric acid electrolyte concept is also applicable to water electrolysis cells, i.e., cells producing both hydrogen and oxygen.

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The Sulfur Cycle Hydrogen Production Plant is based upon a conceptual design and cost estimate originally prepared for NASA (1) and updated for the U. S. Department of Energy (2). The plant is a grass roots, self-sufficient 380 million SCFD hydrogen production unit energized by a very high temperature nuclear reactor (VHTR). Using the updated plant design as a base, revised flow sheets, mass and energy balances, and cost estimates (1976 cost basis) were prepared for plants of the same hydrogen production capacity but with electrolyzer voltages varying between 400 and 800 mV.

Process flow sheets, mass and energy balances, and cost estimates were also prepared for an identically sized grass roots self-sufficient hydrogen production plant using water electrolysis with a sulfuric acid electrolyte. The electrolysis cell configuration, current densities, and cost assumptions are, wherever possible, identical to those for the Sulfur Cycle electrolyzer. The cell voltage and operating temperature are considered parametrically in order to assess the performance objectives of the sulfuric acid cell. The energy for the electrolyzer and its auxiliaries is produced in a VHTR type reactor, operating at the same technological level as for the Sulfur Cycle plant, but configured to produce electric power in a high efficiency Brayton cycle.

The cost of hydrogen production by the Sulfur Cycle water decomposition plant and by the sulfuric acid electrolysis plant are parametrically compared to determine the performance levels at which the two production processes are economically competitive.

Section 3

SULFUR CYCLE HYDROGEN PRODUCTION PROCESS

PROCESS CONCEPT

The water decomposition system used for hydrogen production is the Westinghouse Sulfur Cycle hybrid electrochemical/thermochemical process. The process, in its most general form, consists of two chemical reactions — one for producing oxygen and the other for producing hydrogen. The production of oxygen occurs via the thermal reduction of sulfur trioxide obtained from sulfuric acid.

The equilibrium for Reaction 1 lies to the right at temperatures above 1000K. Catalysts are available for accelerating the rate of sulfur trioxide reduction to sulfur dioxide and oxygen. The process is completed by using the sulfur dioxide from the thermal reduction step to depolarize the anode of a water electrolyzer. The overall reaction occurring electrochemically is

This is comprised of the individual reactions:

The net results of Reactions 1 and 2 is the decomposition of water into hydrogen and oxygen. Sulfur oxides are involved as recycling intermediates. Although electrical power is required in the electrolyzer, much smaller quantities than those necessary in conventional electrolysis are needed. The theoretical voltage to decompose water is 1.23 V, with many commercial electrolyzers requiring over 2.0 V. The power requirements for Reaction 2 (0.17 volts at unit activity for reactants and products) are thus seen to theoretically be less than 15 percent of those required in conventional electrolysis. This dramatically changes the heat and work required to decompose water and leads to improved thermal efficiencies.

The process is shown schematically in Figure 3-1. Hydrogen is generated electrolytically in an electrolysis cell which anodically oxidizes sulfurous acid to sulfuric acid while simultaneously generating hydrogen at the cathode. Sulfuric acid formed in the electrolyzer is then vaporized, using thermal energy from a high temperature heat source. The vaporized sulfuric acid (sulfur trioxide-steam mixture) flows to an indirectly heated reduction reactor where sulfur dioxide and oxygen are formed. Wet sulfur dioxide and oxygen flow to a separation system, where oxygen is produced as a process co-product and the sulfur dioxide is recycled to the electrolyzer.

The cycle has the potential for achieving high thermal efficiencies while using common and inexpensive chemicals. The product hydrogen and oxygen streams are available under pressure and at high purity. As a result, they may be pipelined and used without detrimental environmental effects and without jeopardizing processes which employ the gas.

TECHNOLOGY STATUS

Development work on the Sulfur Cycle Water Decomposition System was started in 1973 by the Westinghouse Electric Corporation with its own funding. Funding assistance from the Department of Energy (or its predecessor organizations) was first made available in 1976. The development activities are currently underwritten by the Department of Energy and Westinghouse.

The overall development program plan is to achieve, by 1983, the milestone of an operating bench scale process development unit (PDU). This program is discussed in Appendix B, Development Requirements, of this report. The most recent published results of the work already done are in Reference 2.

The program is structured to solve the various technological problems in the process steps shown in Figure 3-1. The electrolysis tasks call for work on anodes, cathodes, membranes, catalysts, fabrication techniques, and cell performance over a growing scale from single cell atmospheric pressure test units to large multicell pressurized electrolyzers. A cell performance level of 600 mV, when operating at 200 mA/cm^2 current density, 50°C , atmospheric pressure, and a sulfuric acid concentration of 50 percent has already been demonstrated in the laboratory. The objective of the program is to achieve this same performance at operating parameters up to 80 percent acid concentration, 100°C , and 20 atmosphere operating pressure.

Figure 3-1. Hydrogen Generation Schematic Diagram

The development areas related to the sulfuric acid vaporization step are completely dedicated to the evaluation of materials for containment of high pressure, high temperature, boiling sulfuric acid. Preliminary materials screening tests have shown several non-noble metals which show promise of being able to meet the desired operating conditions.

In the area of oxygen generation, two tasks are being undertaken. One is to evaluate catalysts that promote the high temperature reaction in order to find one that is economically optimum for the process, i.e., low cost, long life, and high activity. Several catalysts have already been evaluated; engineering judgement must await completion of the planned tests and economic trade-off studies to assess the impact of the alternate catalysts. The oxygen production step, operating at temperatures to about 1600°F, also requires a materials evaluation task to determine suitable materials for containing very high temperature steam, SO₃, SO₂, and O₂, on one surface, and a helium heat source fluid on the other. Materials screening tests are currently underway.

The oxygen recovery, or separation, step of the process is one requiring engineering evaluation rather than development. Several processes can afford the required separations. Further evaluation of process flow sheets will determine the most desirable option.

In addition to the technology development aspects of the program, work is underway on the design of a laboratory model, to be operational by the end of 1978, in which all the steps of the Sulfur Cycle will be put together in a closed cycle configuration producing two liters per minute of hydrogen. This will demonstrate that the process can operate in a cyclic manner, with re-use of all reactants.

PROCESS PLANT CONCEPTUAL DESIGN

General

The hydrogen production plant, as in References 1 and 2, is based on the hybrid Sulfur Cycle process for decomposing water, driven by a very high temperature nuclear reactor (VHTR) that provides an 1850°F helium working gas for electric power production and process heat. The plant is sized to produce 380 million SCFD of electrolytically pure hydrogen at a pressure of 300 psi. The plant is designed

to be completely energy self-sufficient, i.e., no energy source is used by the plant other than the energy produced by the nuclear fuel in the VHTR, and no energy form leaves the plant other than the hydrogen product and the low temperature heat dissipated through the plant's cooling tower. To account for the uncertainties in the ultimate technological capability of the hydrogen production process, three different operating voltages of the electrolyzer are assumed, as follows:

- Case 1 - 400 mV
- Case 2 - 600 mV
- Case 3 - 800 mV

Plant Design

The process flow sheet for the Sulfur Cycle is shown in Figure 3-2. Table 3-1 shows the temperature, pressure, flow rate and composition for each stream in Figure 3-2 when the electrolyzer is operating at 600 mV (Case 2). For the other cell operating voltage conditions (Cases 1 and 3), the flow streams are the same as Case 2 except for those streams shown in Table 3-2. There is no power generation in the plant for purposes other than on-site consumption. The VHTR provides all the thermal and electrical energy requirements for the entire plant, and ultimate heat dissipation is through a cooling tower. In addition to the production of electricity, the power plant includes mechanical drive steam turbines directly connected to the helium circulators of the VHTR intermediate heat transfer loop. Helium circulators in the VHTR primary loops are either electric motor driven (very high temperature IXH loops) or helium gas turbine driven (high temperature IHX loops).

The electrolyzer design concept is shown in Figure 3-3. It is a bi-polar arrangement of pressed carbon flow through electrodes, based upon the experimental work performed on laboratory electrolyzers using porous carbon anodes and cathodes. The design is based upon the production of 80 weight percent sulfuric acid in the electrolyzers with the subsequent decomposition of the acid at 1600°F and 300 psia. The hydrogen is generated in the electrolyzer and leaves the process plant at a pressure of 300 psia. The expansion of the oxygen stream through a turbine prior to its venting provides some of the plant's auxiliary power.

Referring to the flow sheet of Figure 3-2, sulfuric acid produced in the electrolyzer, stream 61, is sent to a flash drum, FD-1, where the pressure is reduced from

NOMENCLATURE	
VHTR HEAT SOURCE	
A	ANOLYTE
AV 1,2	ACID VAPORIZERS
C	CATHOLYTE
C 1	COMPRESSOR
CDT 1,2	CIRCULATOR DRIVE TURBINES
CW	COOLING WATER
DR 1,2	DECOMPOSITION REACTORS
E 1,2,3,4,5,6,7	HEAT EXCHANGERS
EL	ELECTROLYZERS
FD 1	FLASH DRUM
GTG 1	GAS TURBINE GENERATOR
K 1,2,3,4,5,6	KNOCK-OUT DRUMS
MD 1	MIXING DRUM
OH 1	OXYGEN HEATER
P 1,2,3,4,5,6,7,8	PUMPS
PH 1,2	PRE HEATERS
SC 1	STEAM CONDENSER
SG 1,2	STEAM GENERATORS
SS 1	STEAM SUPERHEATER
SR 1	STEAM REHEATER
ST 1,2,3,4,5,6,7	SURGE TANKS
STG 1	STEAM TURBINE GENERATOR
T 1,2,3	TOWERS

Figure 3-2. Sulfur Cycle Flowsheet

Table 3-1

FLOW SHEET CONDITIONS - CASE 2 (0.6 VOLT CASE)

Stream No	1	2	3	4	5	6	7	8	9
Temperature, °F	80	90	261	261	261	90	90	261	621
Pressure, psia (1)	300	75	75	75	75	75	300	300	300
Flow Rate, lb/hr (2)	753 9	8065 5	8065 5	7880 4	185 0	185 0	462 4	7880 4	7800 4
Composition, Wt Percent									
H ₂									
O ₂									
H ₂ O	100 0	19 5	19 5	20 0				20 0	20 0
SO ₂		2 3	2 3		100 0	100 0	100 0		
SO ₃									
H ₂ SO ₄		78 2	78 2	80 0				80 0	80 0
Stream No	10	11	12	13	14	15	16	17	18
Temperature, °F	732	846	846	846	1600	1500	1564	981	711
Pressure, psia (1)	300	300	300	300	300	300	300	300	300
Flow Rate, lb/hr (2)	7880 4	7880 4	5013 2	2867 2	5013 2	2867 2	7880 4	7880 4	3426 2
Composition, Wt Percent									
H ₂									
O ₂					8 5	8 5	8 5	8 5	8 5
H ₂ O	20 0	24 2	24 2	24 2	34 7	34 7	34 7	34 7	29 6
SO ₂					34 0	34 0	34 0	34 0	34 0
SO ₃		18 7	18 7	18 7	22 8	22 8	22 8	22 8	
H ₂ SO	80 0	57 1	57 1	57 1					27 9

Notes: (1) Pressure Values are Nominal

(2) Flow in 10³ lb/hr

Table 3-1 (Cont'd)

FLOW SHEET CONDITIONS - CASE 2 (0.6 VOLT CASE)

Stream No.	19	20	21	22	23	24	25	26	27
Temperature, °F	711	711	711	657	86	171	171	333	171
Pressure, psia (1)	300	300	300	300	15	300	300	300	300
Flow Rate, lb/hr (2)	4454.3	158.9	4295.3	669.5	669.5	158.9	4295.3	3426.2	3426.2
Composition, Wt. Percent									
H ₂									
O ₂	8.5	8.5	8.5	100.0	100.0	8.5	8.5	8.5	8.5
H ₂ O	29.6	29.6	29.6			29.6	29.6	29.6	29.6
SO ₂	34.0	34.0	34.0			34.0	34.0	34.0	34.0
SO ₃									
H ₂ SO ₄	27.9	27.9	27.9			27.9	27.9	27.9	27.9
Stream No.	28	29	30	31	32	33	34	35	36
Temperature, °F	171	171	171	171	171	171	171	171	102
Pressure, psia (1)	300	300	300	300	300	300	300	300	300
Flow Rate, lb/hr (2)	1456.7	1969.5	67.0	91.9	1826.6	2468.7	3350.3	4530.1	3350.3
Composition, Wt. Percent									
H ₂									
O ₂	20.0		20.0		20.0		20.0		20.0
H ₂ O		51.4		50.8		51.5		51.4	
SO ₂	80.0		80.0		80.0		80.0		80.0
SO ₃									
H ₂ SO ₄		48.6		49.2		48.5		48.6	

Notes: (1) Pressure Values are Nominal

(2) Flow in 10³ lb/hr

Table 3-1 (Cont'd)

FLOW SHEET CONDITIONS - CASE 2 (0.6 VOLT CASE)

Stream No.	37	38	39	40	41	42	43	44	45
Temperature, °F	102	102	68	68	68	102	68	68	68
Pressure, psia (1)	300	300	300	300	300	300	300	300	300
Flow Rate, lb/hr (2)	1191.7	2158.6	1218.5	276.7	941.9	2435.2	4530.1	684.5	4787
Composition, Wt. Percent									
H ₂									
O ₂	56.2		56.2		72.7			100.0	
H ₂ O							51.4		48.7
SO ₂	43.8	100.0	43.8	100.0	27.3	100		Trace	5.4
SO ₃									
H ₂ SO ₄							48.6		45.9
Stream No	46	47	48	49	50	51	52	53	54
Temperature, °F	78	102	78	78	90	90	319	102	102
Pressure, psia (1)	300	300	300	300	75	75	300	300	300
Flow Rate, lb/hr (2)	753.9	2483.7	669.5	15.0	2565.0	75.3	75.3	26.8	48.5
Composition, Wt. Percent									
H ₂									
O ₂			100.0	100.0		19.9	19.9	56.0	
H ₂ O	100.0				20.0				
SO ₂	Trace	100.0				80.1	80.1	44.0	100.0
SO ₃									
H ₂ SO ₄					80.0				

Notes: (1) Pressure Values are Nominal

(2) Flow in 10³ lb/hr

Table 3-1 (Cont'd)

FLOW SHEET CONDITIONS - CASE 2 (0.6 VOLT CASE)

Stream No.	55	56	57	58	59	60	61	62	63
Temperature, °F	90	100	99	96	110	100	100	90	90
Pressure, psia (1)	75	300	300	300	300	300	300	75	75
Flow Rate, lb/hr (2)	2565	51303	53868	70241	72723	61754	10969	8065.2	2625.8
Composition, Wt. Percent									
H ₂									
O ₂									
H ₂ O	20.0	20.0	20.0	21.0	19.0	19.0	19.0	19.5	19.5
SO ₂				8.7	4.8	4.8	4.8	2.3	2.3
SO ₃									
H ₂ SO ₄	80.0	80.0	80.0	70.2	76.2	76.2	76.2	78.2	78.2
Stream No.	64	65	66	67	68	69	70	71	72
Temperature, °F	100	90	850	479	479	850	115	479	115
Pressure, psia (1)	300	75	1300	225	225	205	1.47	225	1.47
Flow Rate, lb/hr (2)	84.36	277.4	4330	3607	3607	3607	646.6	646.6	646.4
Composition, Wt. Percent									
H ₂	100.0								
O ₂									
H ₂ O			100.0	100.0	100.0	100.0	100.0	100.0	100.0
SO ₂		100.0							
SO ₃									
H ₂ SO ₄									

Notes: (1) Pressure Values are Nominal

(2) Flow in 10³ lb/hr

Table 3-1 (Cont'd)

FLOW SHEET CONDITIONS - CASE 2 (0.6 VOLT CASE)

Stream No.	73	74	75	76	77	78	79	80	81
Temperature, °F	115	116	400	577	470	171	1700	925	771
Pressure, psia (1)	1.47	400	1300	1300	300	300	990	975	960
Flow Rate, lb/hr (2)	3645	3645	3645	3645	4295.3	4295.3	3763	3763	3763
Composition, Wt. Percent									
H ₂									
O ₂					8.5	8.5			
H ₂ O	100.0	100.0	100.0	100.0	29.6	29.6			
SO ₂					34.0	34.0			
SO ₃									
H ₂ SO ₄					27.9	27.9			
He							100.0	100.0	100.0
Stream No.	82	83	84	85	86				
Temperature, °F	627	1400	925	848	627				
Pressure, psia (1)	945	645	630	615	600				
Flow Rate, lb/hr (2)	3763	6227	6227	6227	6227				
Composition, Wt. Percent									
H ₂									
O ₂									
H ₂ O									
SO ₂									
SO ₃									
H ₂ SO ₄									
He	100.0	100.0	100.0	100.0	100.0				

Notes: (1) Pressure Values are Nominal

(2) Flow in 10³ lb/hr

Table 3-2

FLOW RATE AND TEMPERATURE DIFFERENCES BETWEEN CASE 1, 2 AND 3*

<u>Stream No.</u>	<u>Item</u>	<u>Case 1</u>	<u>Case 2</u>	<u>Case 3</u>
66	Flow Rate (10^3 lb/hr) Temperature ($^{\circ}$ F)	3166 850	4330 850	5447 850
67	Flow Rate (10^3 lb/hr) Temperature ($^{\circ}$ F)	3166 479	4330 479	5447 479
68	Flow Rate (10^3 lb/hr) Temperature ($^{\circ}$ F)	2597 479	3607 479	4525 479
69	Flow Rate (10^3 lb/hr) Temperature ($^{\circ}$ F)	2597 850	3607 850	4525 850
70	Flow Rate (10^3 lb/hr) Temperature ($^{\circ}$ F)	2597 115	3607 115	4525 115
73	Flow Rate (10^3 lb/hr) Temperature ($^{\circ}$ F)	3166 115	4330 115	5447 115
74	Flow Rate (10^3 lb/hr) Temperature ($^{\circ}$ F)	3166 116	4330 116	5447 116
75	Flow Rate (10^3 lb/hr) Temperature ($^{\circ}$ F)	3166 400	4330 400	5447 400
76	Flow Rate (10^3 lb/hr) Temperature ($^{\circ}$ F)	3166 577	4330 577	5447 577
78	Flow Rate (10^3 lb/hr) Temperature ($^{\circ}$ F)	4295 280	4295 171	4295 171
81	Flow Rate (10^3 lb/hr) Temperature ($^{\circ}$ F)	3763 815	3763 771	3763 734
83	Flow Rate (10^3 lb/hr) Temperature ($^{\circ}$ F)	4897 1400	6227 1400	7943 1400
84	Flow Rate (10^3 lb/hr) Temperature ($^{\circ}$ F)	4897 860	6227 975	7943 1067
85	Flow Rate (10^3 lb/hr) Temperature ($^{\circ}$ F)	4897 742	6227 848	7943 942
86	Flow Rate (10^3 lb/hr) Temperature ($^{\circ}$ F)	4897 627	6227 627	7943 627

*All other flow streams characteristics are identical regardless of cell voltage.

Case 1 - 400 mV

Case 2 - 600 mV

Case 3 - 800 mV

Figure 3-3. Sulfur Cycle Hybrid System

300 to 75 psi. This outgases much of the dissolved SO_2 which is in this stream. The SO_2 is condensed in the heat exchanger E-2 before being returned to the mixing drum, MD-1. The acid stream leaving FD-1 is split, with a portion being sent to the stripping tower, T-1, where a clean oxygen stream is used to strip the remaining dissolved SO_2 . The resultant SO_2 -oxygen mixture is compressed, with compressor C-1, and returned to the SO_2 -oxygen separation system. The SO_2 -free sulfuric acid (stream 50) is added to the catholyte recirculation system through surge tanks ST-6 and ST-7 and replenishes the losses resulting from the differential pressure maintained between the cell catholyte and anolyte. The remaining acid from FD-1 is sent to the thermal decomposition system.

In the thermal decomposition system, the acid is first heated to 261°F in preheater PH-1 and then sent to surge Tank ST-2, where dissolved SO_2 is removed and recirculated to MD-1. The SO_2 -free acid is then heated to its boiling point in preheater 2 and AV-1 and vaporized in AV-2. The gases leaving AV-2 consist of a mixture of H_2SO_4 , sulfur trioxide, and steam. This is sent to the SO_3 thermal decomposition reactor, shown as a split reactor in DR-1-1 and DR-1-2. Approximately 64 percent of the incoming feed to the decomposition system is sent to DR1-1. This reactor is heated by the process heat source and decomposes SO_3 at temperatures up to 1600°F . The remaining part of the acid stream is sent to DR1-2, which is recuperatively heated by the off gases from both DR1-2 and DR1-1. A gas phase mixture of steam, unreacted SO_3 , SO_2 , and oxygen leaves the decomposition reactors at a temperature of about 980°F and is recuperatively cooled against the incoming acid streams in AV-1, PH-2, and PH-1.

The stream leaving AV-1 is split into two streams. One of these continues to preheat incoming acid via PH-2 and PH-1, while the other goes in parallel to the oxygen preheater OH-1 and the steam cycle for electric power production. The oxygen preheater heats oxygen from 78°F , the temperature at which it leaves the stripping tower, to a temperature of 657°F before being expanded in gas turbine GT-1 back to 15 psi and 86°F .

Stream 21 going to the steam cycle is used to heat feedwater and initiate boiling in steam generators SG-1 and SG-2. The energy contained in that stream which cannot be utilized in the power production cycle is rejected, through heat exchanger E-8, to the plant cooling water system and then to the cooling tower.

Following recuperative heat exchange on the off gases from the decomposition reactor system, all the gases (streams 24, 25 and 27) are available at 171°F. Fifty weight percent sulfuric acid is condensed out of this mixture in knockout drums K-1, K-2 and K-3. This acid underflow goes to surge tank 4 where it is collected, cooled with a chilled water coolant, and sent to scrubbing tower T-3. The gas mixture coming from knockout drums K-1, K-2 and K-3 consists of SO_2 and oxygen. These streams are combined and sent to exchanger E-6, where they are cooled to 100°F with cooling water. Bulk SO_2 is condensed, removed in knockout drum K-4, and sent to surge tank 5.

The gases leaving knockout drum 4, stream 37, consist of oxygen and SO_2 . These gases are further cooled in exchanger E-5 to 68°F, and further SO_2 is condensed and removed in knockout drum K-5. This SO_2 joins the SO_2 effluent from K-4 to surge tank 5 and is ultimately recycled via stream 42 back into the mixing drum for the electrolyzer system. The remaining gas mixture from K-5 is oxygen with small amounts of SO_2 . This stream (41) enters the stripping tower, T-3, which is irrigated with a liquor containing the 50 wt percent sulfuric acid removed in knockout drums K-1, K-2 and K-3 which is used to scrub the oxygen stream in the tower. Based upon solubility considerations, substantially all the SO_2 is removed by the liquor in Tower T-3 and flows, via stream 45, back into the mixing drum, MD-1, and ultimately back into the electrolyzer. The gas leaving tower T-3 is oxygen substantially free of all SO_2 , although it is given a precautionary backup scrub in tower T-2 against the incoming process makeup water.

The scrubbed oxygen stream that flows from T-2 is split into two additional streams. One, stream 48, is heated to OH-1 to 657°F before being expanded and either vented to the atmosphere or sold as a co-product. The second, stream 49, goes to tower T-1 and provides the gas required to strip SO_2 from the SO_2 -containing sulfuric acid in that stage.

The very high temperature nuclear reactor (VHTR), suitable for use with the water decomposition system, is predicated on the integration of the technologies from the NERVA nuclear rocket engine program and land-based gas cooled reactor programs into an advanced graphite moderated, helium cooled reactor. The VHTR conceptual design, costs, and R&D program required for demonstration are more fully described in Reference 4. The plant consists of a Nuclear Island producing both very high temperature heat and electric power for the water decomposition

process. The heat is transported to the process via an intermediate heat transfer loop at temperatures sufficiently high to permit peak process temperatures of 1600°F.

The reactor and its coolant loops are contained within a multi-cavity prestressed cast iron reactor vessel (PCIV), as shown in Figure 3-4. The vessel walls contain smaller vertical cavities, or pods, in which are located very high temperature intermediate heat exchangers, circulators, turbogenerators and high temperature intermediate heat exchangers and auxiliary cooling systems for shut-down and emergency cooling of the reactor. Reactor helium coolant enters and discharges from the pods through coaxial piping at the upper end of the cavity, while the intermediate loop, or secondary, helium coolant is introduced and leaves through the bottom of the pod. The PCIV has a continuous internal steel liner to act as a primary coolant boundary and leak-tight membrane. A thermal barrier and insulation system is used to limit the temperature of the liner and minimize the heat loss to the PCIV. A cooling system circulates water through the walls of the PCIV to remove the heat deposited in the vessel. The PCIV is fabricated as a series of foundry cast iron blocks field assembled around the welded steel liner. Prestress cables are wound around the external cylindrical surface imposing the radial and tangential forces required to prevent the castings from separating under the internal gas pressure. Similarly, axial cables running longitudinally, through ports provided in the castings, maintain a compressive stress in the axial direction and carry the axial pressure loads.

The reference reactor core is designed to operate on the uranium-233/thorium-232 cycle, although it could operate equally well on other fuel cycles. The basic concept of fuel moderator blocks for the reactor is similar to that used in other gas cooled reactors. The extruded fuel elements are directly cooled by the helium. An objective in the core thermal design is to use an existing fuel particle; i.e., the TRISO bead, in the fissile fuel element and to achieve a high exit gas temperature without exceeding the fuel particle limitations.

The schematic flow diagram for the VHTR for the three cases are shown in Figures 3-5, 3-6 and 3.7. The reactor provides its energy in a helium stream at a temperature of 1850°F. The primary helium flow is directed to ten loops, four of which are very high temperature intermediate heat exchange (VHTIHX) loops and six of which are high temperature intermediate heat exchange loops (HTIHX).

Figure 3-4. Very High Temperature Reactor (VHTR)

Figure 3-5. VHTR Flow Schematic Case 1 (400 mV Electrolyzer Voltage)

Figure 3-6. VHTR Flow Schematic Case 2 (600 mV Electrolyzer Voltage)

Figure 3-7. VHTR Flow Schematic Case 3 (800 mV Electrolyzer Voltage)

In the VHTIHX loops, heat is transferred to an intermediate helium stream which provides energy, in series, to the decomposition reactor (DR-1-1), the steam reheater (SR-1), and the steam generator (SG-2). The primary helium in the HTIHX loops passes first through a gas turbine before transferring its heat to the intermediate helium loops. The 1400°F intermediate helium coolant from the HTIHX passes, in series, through the acid vaporizer (AV-2), the steam superheater (SS-1), and the steam generator (SG-2).

The electric power production cycle utilizes a steam turbine generator with throttle conditions of 1300 psia and 850°F. Steam is extracted from the exhaust of the high pressure turbine for the helium circulator drive turbines. No steam is extracted for feedwater heating, since "waste heat" from the process provides this energy. Both the low pressure and mechanical drive turbines exhaust at 3 in. Hg. abs.

In Case 1, part of the energy in Stream 21 has to be rejected due to unacceptable "pinch points" in the heat exchangers. In Cases 2 and 3, the heat exchanger E-8 is deleted and all of the energy in Stream 21 is utilized in the steam cycle.

Table 3-3 shows the closed cooling water requirements for the plant for the three cases, including the requirements for the VHTR circulated through a separate tower.

Table 3-4 shows the total electric generation of the plant. This total capacity is utilized in the manner shown in Table 3-5 for each case.

The overall process efficiency, defined as the higher heating value of the hydrogen produced divided by the total thermal energy input to the plant, is shown in Table 3-6 for each case. Figure 3-8 is a plot of overall plant efficiencies versus cell voltage. Note that it is practically a straight line relationship. The desirability of operating at low cell voltage is apparent.

SULFUR CYCLE ECONOMIC EVALUATION

The economics of the 400 mv, 600 mv and 800 mv cases for the Sulfur Cycle plant have been evaluated using 1976 cost levels. The method of estimating used was similar to that employed in the earlier studies (1,2). The economic evaluation also followed, where appropriate, the June 1977 guidelines in the EPRI Technical Assessment Guide. The evaluation considered utility financing, 18 percent annual

Table 3-3
CLOSED COOLING WATER REQUIREMENTS

<u>Heat Source</u>	<u>Cooling Water Flow Rate, GPM</u>		
	<u>Case 1 **</u>	<u>Case 2 **</u>	<u>Case 3 **</u>
VHTR	6,600	6,600	6,600
E-1	490	490	490
E-3	66,470	66,470	66,470
E-4A*	14,400	14,400	14,400
E-6	17,900	17,900	17,900
E-7A*	306	306	306
Chilled Water Refrigerant System	13,500	13,500	13,500
E-8	23,250	0	0
Condenser (SC-1)	<u>304,200</u>	<u>416,550</u>	<u>523,950</u>
TOTAL	447,116	536,216	643,616

* These heat exchangers have two shells, the A shell cooled by the closed cooling water system and the B shell cooled by chilled water.

** Case 1 - 400 mV Electrolyzer Voltage
Case 2 - 600 mV Electrolyzer Voltage
Case 3 - 800 mV Electrolyzer Voltage

Table 3-4
TOTAL ELECTRIC GENERATION

<u>Source</u>	<u>Capacity, KWe</u>		
	<u>Case 1*</u>	<u>Case 2*</u>	<u>Case 3*</u>
Oxygen Expander	24,071	24,071	24,071
Helium Gas Turbines	65,000	81,000	107,000
Steam Turbine Generator	<u>393,110</u>	<u>585,223</u>	<u>767,842</u>
TOTAL	482,181	690,294	898,913

- * Case 1 - 400 mV Electrolyzer Voltage
Case 2 - 600 mV Electrolyzer Voltage
Case 3 - 800 mV Electrolyzer Voltage

Table 3-5
AUXILIARY ELECTRIC POWER REQUIREMENTS

<u>Item</u>	<u>Normally Running Load, KWe</u>		
	<u>Case 1*</u>	<u>Case 2*</u>	<u>Case 3*</u>
VHTR Nuclear Island	32,315	32,315	32,315
Electrolyzers	407,000	610,500	814,020
Electrolyzer Auxiliary Pumps (P-1,2,3)	2,296	2,296	2,296
SO ₃ Reduction System Pumps (P-4,5)	1,140	1,140	1,140
SO ₂ /O ₂ Separation System Pump (P-7)	80	80	80
Gas Compressors (C-1)	1,015	1,015	1,015
Ammonia Compressors (Chilled H ₂ O Systems)	15,000	15,000	15,000
Chilled Water Circu- lating Pumps	160	160	160
Boiler Feed Pumps (P-6,8)	5,145	7,034	8,849
Circulating Water Pumps	13,665	16,389	19,673
Water Make-Up, Etc.	365	365	365
Miscellaneous	<u>4,000</u>	<u>4,000</u>	<u>4,000</u>
TOTAL	482,181	690,294	898,913

- * Case 1 - 400 mV Electrolyzer Voltage
Case 2 - 600 mV Electrolyzer Voltage
Case 3 - 800 mV Electrolyzer Voltage

Table 3-6

OVERALL PROCESS EFFICIENCY
SULFUR CYCLE HYDROGEN PRODUCTION

<u>Item</u>	<u>Case 1*</u>	<u>Case 2*</u>	<u>Case 3*</u>
Energy Output (Btu/hr) (380×10^6 SCFD of H_2 at 325 Btu/SCF)	5.146×10^9	5.146×10^9	5.146×10^9
VHTR Rating (MWt)	2,839	3,221	3,720
Energy Input (Btu/hr)	9.687×10^9	10.991×10^9	12.694×10^9
Efficiency (%)	53.1	46.8	40.5

- * Case 1 - 400 mV Electrolyzer Voltage
 Case 2 - 600 mV Electrolyzer Voltage
 Case 3 - 800 mV Electrolyzer Voltage

Figure 3-8. Sulfur Cycle Overall Plant Efficiency Versus Cell Voltage

charge on depreciating investment, an 80 percent capacity factor, and no credit for the oxygen produced.

The capital cost estimate is based on preliminary sizing of most of the major plant equipment and determining appropriate costs for that equipment. Factors, based on experience with these types of systems, were used to account for the costs of installation, piping, valves, instrumentation, structures, and miscellaneous equipment. The code of accounts used to assure that all costing elements were accounted for included, for the hydrogen production plant, the On-sites and the Off-sites. The On-sites consist of:

- Battery F, Electrolyzer Power Supply - (Regulated D. C. Power to the Electrolyzer).
- Battery G, Electrolyzers and Auxiliaries
- Battery H, Sulfuric Acid Decomposition
- Battery I, SO_2/O_2 Separation
- Battery J, Turbine Generation System

The Off-Sites consist of:

- Closed Cooling Water System
- Make-up and Feedwater System
- Waste Water System
- Chilled Water System
- Electrical Auxiliary System
- Site, Site Improvements and General Facilities

The VHTR costs used in the economic evaluation were taken from Reference 2 and adjusted for different ratings and interface conditions.

Table 3-7 shows the estimate of the on-sites and off-sites direct capital cost for the Sulfur Cycle hydrogen production plant. Note that the cost of the Very High Temperature Nuclear Reactor (VHTR) heat source is not included here. The estimated cost of Battery G, Electrolyzer and Auxiliaries, is further detailed in Table 3-8, so that a comparison can be more readily drawn between these costs and the comparable costs for the sulfuric acid electrolysis system (Table 4-7).

The increases in the direct capital cost, as cell voltage increases, are in the electrolyzer power supply (Battery F), turbine generation system (Battery J), and the off-sites cooling water system and feedwater make-up. These account for the fifty million dollars cost difference between the 400 mV and 800 mV cases. Since the performance characteristics of the thermal decomposition portion of the plant do not change with cell voltage (fixed hydrogen production rate of 380 million SCFD), the direct capital costs of Batteries H and I are the same for each of the voltage cases. A plot of the total direct capital cost for the hydrogen production plant is shown in Figure 3-9.

The total plant investment is given in Table 3-9 and includes the direct costs of the hydrogen production plant and the VHTR heat source. The nondepreciating assets are land and land rights and are shown separately from the other direct costs for the purpose of evaluation. The special materials under depreciating assets comprise the initial supply of coolant, chemicals, catalysts, lubricants, and other materials needed for operation of the overall plant. A contingency of 20 percent is applied to both the hydrogen generation facilities and the VHTR physical plant. This is slightly more conservative than in Reference 2, where a 15 percent contingency was used for the VHTR and hydrogen generation facilities.

Indirect costs in Table 3-9 are expense items of a general nature which apply to the overall project of building an operable plant, rather than to one of the direct costs. These costs, except for interest during construction, have not been estimated in detail but calculated as a percentage of the direct cost, as in Reference 2. The indirect costs for the water decomposition facilities are calculated as incremental costs to that already included in the VHTR.

Table 3-7

SULFUR CYCLE PLANT
DIRECT CAPITAL COSTS - 1976 BASIS

<u>Electrolyzer Voltage Cases</u>	<u>Case 1*</u> <u>(0.4 Volts)</u>		<u>Case 2*</u> <u>(0.6 Volts)</u>		<u>Case 3*</u> <u>(0.8 Volts)</u>	
	<u>\$10³</u>	<u>\$/kW⁽²⁾</u>	<u>\$10³</u>	<u>\$/kW⁽²⁾</u>	<u>\$10³</u>	<u>\$/kW⁽²⁾</u>
<u>On Site</u>						
Battery F - Electrolyzer Power Supply	\$ 17,622	\$ 11.7	\$ 25,899	\$ 17.2	\$ 34,187	\$ 22.7
Battery G - Electrolyzer & Aux.	169,211	112.2	170,978	113.4	172,745	114.5
Battery H - Sulfuric Acid Decomposition	144,800	96.0	144,800	96.0	144,800	96.0
Battery I - SO ₂ /O ₂ Separation	20,420	13.5	20,420	13.5	20,420	13.5
Battery J - Turbine Generation System	<u>49,717</u>	<u>33.0</u>	<u>58,776</u>	<u>39.0</u>	<u>74,226</u>	<u>49.2</u>
On-Sites Sub-Total	\$401,770	\$266.4	\$420,873	\$279.1	\$446,378	\$296.0
<u>Off-Sites</u>						
Cooling Water System & Water Intake	\$ 9,280	\$ 6.2	\$ 10,254	\$ 6.8	\$ 11,154	\$ 7.4
Makeup & Feedwater	7,769	5.2	8,009	5.3	8,237	5.5
Waste Water Treatment	340	0.2	340	0.2	340	0.2
Chilled Water System	3,085	2.0	3,085	2.0	3,085	2.0
Electrical Aux. Power	24,434	16.2	25,979	17.2	27,619	18.3
Land & Land Rights(1)	200	0.1	200	0.1	200	0.1
Site Improvements & Facilities	1,080	0.7	1,080	0.7	1,080	0.7
Admin./Service/Lab Bldgs.	1,430	0.9	1,430	0.9	1,430	0.9
Instrument & Plant Air	2,650	1.8	2,650	1.8	2,650	1.8
Maintenance Facilities	1,550	1.0	1,550	1.0	1,550	1.0
Fire Protection, Communication	950	0.6	950	0.6	950	0.6
Furnishings, Fixtures, etc.	<u>250</u>	<u>0.2</u>	<u>250</u>	<u>0.2</u>	<u>250</u>	<u>0.2</u>
Off-Sites Sub-Total	\$ 53,018	\$ 35.1	\$ 55,777	\$ 36.8	\$ 58,545	\$ 38.7
Total Direct Capital Cost	\$454,788	\$301.5	\$ 476,650	\$315.9	\$504,923	\$334.7

(1) Some items in General Off-Site are shared with VHTR and included partially in VHTR costs.

(2) Based on heating value of product hydrogen.

Table 3-8
SULFUR CYCLE PLANT
BATTERY G - ELECTROLYZER AND AUXILIARIES
DIRECT CAPITAL COST - 1976 BASIS
(Thousands of Dollars)

	<u>Case 2 (0.6 Volts)</u>
Electrolyzers	\$144,700
• Cells	\$ 78,500
• Vessel	\$ 35,600
• Internal Structure, etc.	\$ 30,600
Electrolyzer External Supports and Installation	2,300
Electrolyzer Instrumentation	3,700
Electrolyzer Auxiliary Equip- ment, Installed ⁽¹⁾	11,268
D-C Bus Duct, Installed	5,300
Electrolyzer Building	<u>3,710</u>
Total	\$170,978

(¹) Includes installed cost of equipment closely associated with electrolyzer, i.e., heat exchanger E-1, E-2, E-3; pumps P-1, P-2, P-3, P-4; tanks ST-6, ST-7, FD-1, MD-1, T-1, K-6; and compressor C-1 (all shown on Figure 3-2).

Figure 3-9. Sulfur Cycle Plant Direct Capital Cost Versus Cell Voltage - 1976 Basis

Table 3-9
SULFUR CYCLE PLANT
CAPITAL COST SUMMARY - 1976 BASIS
(Thousands of Dollars)

	Case 1* (0.4 Volts)		Case 2* (0.6 Volts)		Case 3* (0.8 Volts)	
	VHTR	H ₂ Prod	VHTR	H ₂ Prod	VHTR	H ₂ Prod
<u>Non-Depreciating Assets</u>						
Land & Land Rights	\$ 800	\$ 200	\$ 800	\$ 200	\$ 800	\$ 200
<u>Depreciating Assets</u>						
Special Materials	350	1,170	350	1,170	350	1,170
Physical Plant	241,268	454,588	260,252	476,450	283,743	504,723
Sub-Total	241,618	455,758	260,602	477,620	284,093	505,893
Contingency (20%)	48,324	91,152	52,120	95,524	56,819	101,179
Sub-Total	289,942	546,910	312,722	573,144	340,912	607,072
Indirect Costs						
Construction Fac., Equip., Services	18,730	11,485	20,202	12,036	22,023	12,749
Engineering Serv.	45,521	19,142	49,097	20,060	53,523	21,248
Other Costs	14,439	4,922	15,574	5,158	16,977	5,464
Interest During Constr. ⁽¹⁾ (8% Yr.)	<u>100,552</u>	<u>94,068</u>	<u>108,452</u>	<u>98,581</u>	<u>118,228</u>	<u>104,416</u>
Total Depreciating	469,184	676,527	506,047	708,979	551,663	750,949
Total Plant Investment	<u>\$469,984</u>	<u>\$676,727</u>	<u>\$506,847</u>	<u>\$709,179</u>	<u>\$552,463</u>	<u>\$751,149</u>
	\$1,146,711		\$1,216,026		\$1,303,612	
	(\$760/kw) ²		(\$806/kw) ²		(\$864/kw) ²	

(1) Eight-Year Construction Schedule

(2) Based on Heating Value of Product Hydrogen

The construction facilities, equipment, and services include general costs associated with the plant construction. These indirect costs include such items as field offices, warehouses, temporary power and utility lines, cost or rental of construction equipment and supplies, purchase of electric power, and water and other utilities, security guards, training programs for the labor force, inspection and testing of construction materials, site cleanup, insurance and any other general cost associated with plant construction.

The engineering services include items such as preliminary investigations; site selection; air and water environmental studies; subsurface investigations; preparation of specifications and evaluation of proposals for major equipment packages, preparation of preliminary and final design documents, design reviews, procurement, inspection, and expediting of materials and equipment; preparation of pre-operational test and plant startup procedures; assistance in securing plant permits; management and direction of construction activities, including selection of subcontractors, scheduling maintaining cost and quality control; on-site procurement and receiving of materials and equipment; field accounting; supervising and pre-operational testing of systems and components; field engineering inspection of construction work to assure compliance with plans and specifications; and preparation of as-built drawings.

Other costs include the owner's property and all-risk insurance, state and local property taxes on the site and improvements during construction, sales taxes on purchased materials and equipment, staff training, plant startup, and the owner's general and administrative (G&A) costs.

Interest during construction is calculated as simple interest, at an eight percent annual rate, on the plant investment as it is made. For the purpose of the evaluation, it is assumed that the land is purchased six months prior to the start of the project and that special materials are delivered and paid for nine months prior to plant commercial operation. The remainder of the plant investment is made as design and construction proceeds. Figure 3-10 shows the rate of expenditures as a function of time. The overall project period of eight years is dictated by the expected future design, licensing, and construction time for the VHTR.

Figure 3-10. Nuclear Water Decomposition - Plant Capital Investment Expenditures Versus Time

A summary plot of the sulfur cycle total capital cost is shown in Figure 3-11 for the cell voltage cases ranging from 0.4 volts to 0.8 volts. Also shown are the contributions of the hydrogen production and the VHTR plant costs.

The sulfur cycle total capital as a function of current density has also been plotted. Figure 3-12 shows the variation in total plant investments for each of the three voltage cases. The curves assume that the voltage shown is achieved at 200 mA/cm^2 . As the current density increases, the voltage is assumed to increase as the result of IR and polarization effects. Therefore, as current density increases, for a constant hydrogen output, the investment tends to decrease due to lower electrolyzer costs and to increase due to the greater electrolyzer power requirements. The net effect is for the total plant investment curve to flatten out at higher current densities.

The hydrogen production cost is given in Table 3-10 and is made up of the contributions of nondepreciating and depreciating capital, operation and maintenance, and nuclear fuel cycle costs. These are calculated on an annual basis. The annual charge on nondepreciating assets; e.g., land, is assumed to be ten percent. The charge on depreciating assets is 18 percent. An 80 percent capacity factor was assumed. Note that in Table 3-10, a nuclear fuel cost of $50\text{¢}/10^6 \text{ Btu}$ is used and that no credit is taken for the oxygen produced. Figure 3-13 shows the variation of hydrogen "gate price" with cell voltage.

To account for the variability of the hydrogen "gate price" with changes in nuclear fuel cost, additional analyses are performed. Figure 3-14 shows the hydrogen production cost plotted against nuclear fuel cost. It is interesting to note that by doubling the nuclear fuel cost, the hydrogen "gate price" increases only 13.5 percent for the worst voltage case (0.8 volts).

The effect of current density on hydrogen "gate price" was also evaluated. Figure 3-15 gives the hydrogen production cost for the 0.6 volt case versus current density for nuclear fuel costs of $\$0.50/10^6 \text{ Btu}$, $\$1.00/10^6 \text{ Btu}$, and $\$1.50/10^6 \text{ Btu}$, respectively. Note that these curves tend to bottom out between 400 mA/cm^2 and 600 mA/cm^2 . For all practicable purposes the hydrogen "gate price" is almost constant. The minimum does, however, seem to shift toward lower current densities as the nuclear fuel cost increases.

Figure 3-11. Sulfur Cycle Plant Capital Cost Summary Variation with Cell Voltage - 1976 Basis

Figure 3-12. Sulfur Cycle Plant Capital Cost Summary Variation with Current Density - 1976 Basis

Table 3-10
SULFUR CYCLE PLANT
HYDROGEN PRODUCTION COST (NO OXYGEN CREDIT)
80% Capacity Factor
Utility Financing
1976 Cost Basis

<u>Electrolyzer Voltage Cases</u>	<u>Case 1</u> <u>(0.4 Volts)</u>	<u>Case 2</u> <u>(0.6 Volts)</u>	<u>Case 3</u> <u>(0.8 Volts)</u>
<u>Annual Costs</u> (Thousands of Dollars)			
Non-Depreciating Capital (1)	\$ 100	\$ 100	\$ 100
Depreciating Capital (2)	206,228	216,705	234,470
Operation & Maintenance	6,373	6,875	7,495
Nuclear Fuel Cycle Cost (3)	<u>33,951</u>	<u>38,519</u>	<u>44,487</u>
Total Annual Cost	\$246,652	\$264,199	\$286,552
Annual H ₂ Production (SCF)	1.11 x 10 ¹¹	1.11 x 10 ¹¹	1.11 x 10 ¹¹
Hydrogen "Gate Price"			
\$ / MSCF	2.22	2.38	2.58
\$ / 10 ⁶ BTU	6.82	7.31	7.93

(1) Land at 10% Annual Charge

(2) 18% Annual Charge

(3) At 50¢/10⁶ BTU for Nuclear
Fuel Cost

Figure 3-13. Sulfur Cycle Plant Hydrogen "Gate Price" Versus Cell Voltage - 1976
Basis ($50¢/10^6$ Btu Nuclear Fuel Cost)

Figure 3-14. Sulfur Cycle Plant Hydrogen "Gate Price" Versus Nuclear Fuel Cost - 1976 Basis

Figure 3-15. Sulfur Cycle Plant Hydrogen "Gate Price" Versus Current Density - 1976 Basis (Nominal 600 mV at 200 mA/cm²)

Section 4.0

SULFURIC ACID ELECTROLYSIS HYDROGEN PRODUCTION PROCESS

PROCESS CONCEPT

Water electrolysis systems using alkaline electrolyzers have been in commercial service for many years. Development activities on water electrolyzers within the United States have been directed towards: 1) improving the performance and economics of alkaline electrolyzers; and 2) the development of the solid polymer electrolyte (SPE) electrolyzers with their high current density capability and potentially low investment cost per unit of production.

The work underway on the Sulfur Cycle Water Decomposition System has resulted in development activities on electrolyzers using a sulfuric acid electrolyte. Recognizing that such an electrolyzer might compare favorably with alternative electrolyzers if used for the direct electrolysis of water, a technoeconomic assessment was conducted.

As a first step in evaluating the potential of sulfuric acid electrolysis, this study considered, for comparison with the Sulfur Cycle hydrogen production system, a water electrolysis plant using, as far as possible, the technology of sulfuric acid electrolyzers being developed under the Sulfur Cycle program.

To make the comparison as consistent as possible, the energy source for the sulfuric acid electrolysis plant was, as for the Sulfur Cycle, a very high temperature nuclear reactor (VHTR). The VHTR is configured, however, for high efficiency electric power production using a helium Brayton Cycle. The electrolysis plant itself uses a process flowsheet typical of many electrolysis plants, modified only to account for the presence of sulfuric acid instead of an alkaline solution.

TECHNOLOGY STATUS

The performance capability of an acid electrolyzer benefits primarily from the higher conductivity of sulfuric acid, as compared to the potassium hydroxide commonly used in alkaline cells. Figure 4-1 shows the conductivity of sulfuric acid and potassium hydroxide as a function of temperature for solutions of maximum conductivity; i.e., about 28 weight percent for both the acid and alkaline solutions.

To estimate the voltage at which an acid electrolyte cell can operate, the general physical characteristics of the cell, as distinguished from a comparable alkaline cell, must be considered. In principle, the acid electrolyzer would have similar electrode spacing and a diaphragm of comparable porosity to the alkaline electrolyzer. The acid electrolyzer requires an inert anode, which can be catalytic, whereas a catalytic anode may be optional in an alkaline system. The concept of the acid electrolyzers used for this study consisted of a bipolar stacking of cells, as shown in Figure 4-2. The cells contain a titanium intercell structure which would be surfaced with the catalytic anode on one side. The titanium would have to be protected on the cathode side, since a cathodic titanium loses its inertness due to oxide reduction and, in fact, is converted to a hydride which decomposes. A lead surfacing is therefore proposed for the cathode side. The membrane can be asbestos or microporous.

The cell voltage was calculated by making appropriate adjustments to the characteristic curve for an alkaline cell. Figure 4-3 shows the resulting voltage for the sulfuric acid electrolysis cell, as a function of current density, as compared to that of an alkaline cell. The alkaline cell voltage curve was taken from a study by Srinivasan and Salzano (5). The sulfuric acid curve represents the improvements to be expected as a result of the improved conductivity of the electrolyte and the presence of a catalytic anode.

A parametric series of curves can also be calculated in a similar fashion to cover a range of cell operating temperatures. Figure 4-4 shows cell voltage versus current density for the sulfuric acid electrolyzer with operating temperatures from 30°C to 150°C. These curves are used to determine the cell voltage for the different operating temperature cases in this study.

Figure 4-1. Conductivity Versus Temperature for KOH and H₂SO₄
(28 Wt.% Solutions)

Figure 4-2. Sulfuric Acid Electrolysis System

Figure 4-3. Calculated Cell Voltage Versus Current Density
for H₂SO₄ Electrolyzer Catalytic Anode

Figure 4-4. Calculated Cell Voltage Versus Current Density for H₂SO₄ Electrolyzer

General

The hydrogen production plant is based on a grass roots self-sufficient water electrolysis process, using a sulfuric acid electrolyte, energized by a very high temperature nuclear reactor (VHTR) configured to produce electric power in a high efficiency Brayton Cycle. The plant production capacity, as for the Sulfur Cycle plant, is 380 million SCFD of hydrogen. The electrolyzer uses a 28 weight percent solution of sulfuric acid as the electrolyte and operates at a pressure of 300 psi. To account for the uncertainties in the ability to predict the operating voltage of the electrolyzer, three different cell voltages, at an operating temperature of 100°C and a current density of 200 mA/cm², were evaluated. These are:

Case 1 - 1.58 Volts

Case 2 - 1.68 Volts

Case 3 - 1.78 Volts

Of these three voltages, Case 2 (1.68 V) represents the calculated voltage for the cell (as shown in Figure 4-3) at the specified temperature, current density, and electrolyte concentration for maximum conductivity (28 weight percent H₂SO₄). Case 2 was, therefore, also used as the base from which the effect of operating temperature on plant performance could be assessed. For this purpose, two more cases are identified. These reflect the same current density as Case 2 and the change in voltage resulting from temperature differences as shown in Figure 4-4. The additional cases are designated:

Case 4 - 1.88 Volts (60°C)

Case 5 - 1.63 Volts (150°C)

Plant Design

The process flow sheet for the sulfuric acid electrolyzer plant is shown in Figure 4-5. Table 4-1 shows, as an example, the temperature, pressure, flow rate, and composition of each stream for Case 2 (1.68 V Case). As can be seen in Figure 4-5, the hydrogen and oxygen gases together with entrained electrolyte leave the cell and move through gas-liquid separators and heat exchangers. The separated electrolyte is recycled through a filter-mixer where makeup water is added. The electrolyte is then returned to the electrolyzer module. The separated gases, which are saturated with water, pass through the water-cooled gas

Figure 4-5. H₂ Production Plant Schematic (1.68 Volt, 100°C Case)

Table 4-1

SUMMARY OF EACH STREAM: CASE 2 (1.68V CASE)

Stream #	Temp. °F	Pressure psia	Mass Flow Rate & Components lbs/hr, g = gas, l = liquid, v = vapor
1	77	300	757,181 H ₂ O (l) high purity feedwater
2	100	300	50,497 H ₂ O (l) from H ₂ & O ₂ coolers
3	74	300	807,678 H ₂ O (l)
4	74	300	807,678 H ₂ O (l) + 4 of 28% H ₂ SO ₄ (l)
5	193	300	47.8 x 10 ⁶ of 28% H ₂ SO ₄ (l)
6	212	300	23,127,532 electrolyte (l) + 669,268 O ₂ (g) + 18,028 H ₂ O (v) + .5 H ₂ SO ₄ (v)
7	212	300	23,609,272 electrolyte (l) + 84,328 H ₂ (g) + 36,056 H ₂ O (v) + .5 H ₂ SO ₄ (v)
8	212	300	23,212,703 electrolyte (l)
9	195	300	23,212,703 electrolyte (l)
10	212	300	23,779,615 electrolyte (l)
11	195	300	23,779,615 electrolyte (l)
12	212	300	669,268 (g) O ₂ + 18,028 H ₂ O (v) + .5 H ₂ SO ₄ (v)
13	100	300	669,268 (g) O ₂ + 1,196 H ₂ O (v) + .5 H ₂ SO ₄ (v)
14	212	300	84,328 (g) H ₂ + 36,056 H ₂ O (v) + .5 H ₂ SO ₄ (v)
15	100	300	84,328 (g) H ₂ + 2,392 H ₂ O (v) + .5 H ₂ SO ₄ (v)
16	100	300	16,833 H ₂ O (l) condensed water
17	77	15	1.775 x 10 ⁶ H ₂ O (l) cooling water
18	97	15	1.775 x 10 ⁶ H ₂ O (l) cooling water
19	77	15	3.433 x 10 ⁶ H ₂ O (l) cooling water
20	97	15	3.433 x 10 ⁶ H ₂ O (l) cooling water
21	77	15	16.776 x 10 ⁶ H ₂ O (l) cooling water
22	97	15	16.776 x 10 ⁶ H ₂ O (l) cooling water

Table 4-1 (Continued)

Stream #	Temp. °F	Pressure psia	Mass Flow Rate & Components
			lbs/hr, g = gas, l = liquid, v = vapor
23	77	15	16.776×10^6 H ₂ O (l) cooling water
24	97	15	16.776×10^6 H ₂ O (l) cooling water
25	97	15	38.748×10^6 H ₂ O (l) cooling water
26	77	15	38.748×10^6 H ₂ O (l) cooling water
27	77	15	6,345,209 H ₂ O (l) make-up cooling water
28	77	15	366,781,791 H ₂ O (l) cooling water
29	77	15	373,127,000 H ₂ O (l) cooling water
30	97	15	334,379,000 H ₂ O (l) cooling water
31	77	15	334,379,000 H ₂ O (l) cooling water
32	97	15	373,127,000 H ₂ O (l) cooling water

coolers and are cooled to 100°F. The condensed water is collected and returned to the makeup feedwater system. The hydrogen gas leaves the electrolyzer at 100°F and 300 psi. The oxygen gas goes to an oxygen heater located in the power plant, where it is heated to 1100°F before entering an oxygen expander where 39.6 megawatts of electrical power are produced. Waste heat is collected by a closed cooling water system and dissipated through a cooling tower. Raw river water is taken to a water-treatment system to provide the makeup cooling water and feedwater of the required quality and quantity for plant operation.

The very high temperature nuclear reactor (VHTR) used to energize the sulfuric acid electrolysis plant is, as in the Sulfur Cycle plant, an advanced graphite moderated helium-cooled reactor capable of outlet coolant temperatures of 1850°F. Unlike the Sulfur Cycle plant, the VHTR is now configured to use the total helium coolant flow in a Brayton Cycle to produce electric power at high thermal efficiencies. Figures 4-6 through 4-10 show the flow schematics of the VHTR Brayton Cycles for the five cases studied. In all cases, it was assumed that a single nuclear reactor would serve the plant, regardless of the fact that the reactor rating for some of the cases exceeds the current nuclear licensing limit of 3800 MWt.

Table 4-2 shows the total electric generation for each of the five cases. The oxygen expander contributes approximately 40 MW(e) to the total power needs of the plant. The mass and energy balances for the electrolysis plant were calculated. Table 4-3 displays the overall energy balance; and Table 4-4 displays the overall mass balance for the Case 2 (1.68 V Case) evaluation.

The overall plant efficiency combines the power generation efficiency and the hydrogen plant efficiency. Table 4-5 shows the power generation efficiency, hydrogen plant efficiency, and overall plant efficiency for each case studied. These efficiencies, as a function of cell voltage at a constant operating temperature of 100°C, are plotted in Figure 4-11. The incentive of operating at lower cell voltages is apparent. Figure 4-12 shows the sulfuric acid electrolyzer overall plant efficiency, at a constant current density of 200 mA/cm^2 , versus the electrolyte temperature. As expected, increasing temperatures result in higher efficiencies, but at a diminishing rate. This is attributed to the fact

T = TEMPERATURE, °F

P = PRESSURE, psia

W = FLOW RATE, 10⁶ LB/HR

Figure 4-6. VHTR Flow Schematic (Case 1, 1.58 Volt, 100°C)

T = TEMPERATURE, °F

P = PRESSURE, psia

W = FLOW RATE, 10^6 LB/HR

Figure 4-7. VHTR Flow Schematic (Case 2, 1.68 Volt, 100°C)

Figure 4-8. VHTR Flow Schematic (Case 3, 1.78 Volt, 100°C)

T = TEMPERATURE, °F

P = PRESSURE, psia

W = FLOW RATE, 10^6 LB/HR

Figure 4-9. VHTR Flow Schematic (Case 4, 1.88 Volt, 60°C)

T = TEMPERATURE, °F

P = PRESSURE, psia

W = FLOW RATE, 10⁶ LB/HR

Figure 4-10. VHTR Flow Schematic (Case 5, 1.63 Volt, 150°C)

Table 4-2
TOTAL ELECTRIC GENERATION

<u>Source</u>	Capacity, MW(e)				
	<u>Case 1</u>	<u>Case 2</u>	<u>Case 3</u>	<u>Case 4</u>	<u>Case 5</u>
Oxygen Expander	40	40	40	40	40
Helium Gas Turbine	<u>1567</u>	<u>1669</u>	<u>1771</u>	<u>1872</u>	<u>1618</u>
TOTAL	1607	1709	1811	1912	1658

NOTE: Case 1 - Electrolyzer Voltage 1.58 V; Temperature 100°C
Case 2 - Electrolyzer Voltage 1.68 V; Temperature 100°C
Case 3 - Electrolyzer Voltage 1.78 V; Temperature 100°C
Case 4 - Electrolyzer Voltage 1.88 V; Temperature 60°C
Case 5 - Electrolyzer Voltage 1.63 V; Temperature 150°C

Table 4-3

OVERALL ENERGY BALANCE FOR THE PRODUCTION OF 380×10^6 SCFD
HYDROGEN (CASE 2 - 1.68 V, 100°C)

<u>ITEM</u>	<u>ENERGY (MEGAWATTS)</u>	
	<u>INPUT</u>	<u>OUTPUT</u>
Electrolyzer		
Water Enthalpy	9.99	-
Power Input	1,705.55	-
Hydrogen Enthalpy	-	1.93
Oxygen Enthalpy	-	1.04
Decomposition Enthalpy	-	1,588.00
Heat Loss	-	124.57
Sub-Total	1,715.54	1,715.54
Pump Loads		
Electrolyzer	3.50	2.98
Power Plant	16.86	14.33
Heat Loss	-	3.05
Sub-Total	20.36	20.36
Cooling Towers		
Water Enthalpy	7,106.03	4,840.62
Make-Up Enthalpy	83.74	-
Air Enthalpy	3,264.69	5,568.00
Heat Loss	-	45.84
Sub-Total	10,454.46	10,454.46
Total	12,190.36	12,190.36

Table 4-4

OVERALL MASS BALANCE FOR THE PRODUCTION OF 380×10^6 SCFD OF H_2
(CASE 2 - 1.68 V, 100°C)

<u>ITEM</u>	<u>INPUT</u>	<u>OUTPUT</u>
	<u>Lbs/Hr</u>	<u>Lbs/Hr</u>
Electrolyzers (1)		
Feed Water	757,184	- - -
Sulfuric Acid (2)	1	1
Hydrogen (300 psia)	- - -	84,328
Oxygen (300 psia)	- - -	669,268
Water In H_2 and O_2	- - -	3,588
Subtotal:	757,185	757,185
Cooling Towers		
Water From Electrolyzer	38,748,000	- - -
Water From Power Plant	334,379,000	- - -
Make-up Water	6,345,209	- - -
Air (Dry Basis)	341,691,000	341,691,000
Water Vapor In Air	4,100,292	10,445,501
Cooling Water	- - -	373,127,000
Subtotal:	725,263,501	725,263,501
TOTAL:	726,020,686	720,020,686

(1) An initial charge of 13.4×10^6 lbs of sulfuric acid in 34.4×10^6 lbs of water is retained in the system.

(2) Sulfuric acid entrained in H_2 and O_2 off gases.

Table 4-5
OVERALL PROCESS EFFICIENCY
SULFURIC ACID ELECTROLYSIS PLANT

	Case 1*	Case 2*	Case 3*	Case 4*	Case 5*
Energy Output (Btu/Hr) (380×10^6 SCFD of H_2 at 325 Btu/SCF)	5.146×10^9	5.146×10^9	5.146×10^9	5.146×10^9	5.146×10^9
VHTR Rating (MWt)	3474	3699	3925	4143	3591
Energy Input (Btu/Hr)	11.85×10^9	12.62×10^9	13.39×10^9	14.14×10^9	12.25×10^9
Efficiency (%)	43.4	40.8	38.4	36.4	42.0

- * Case 1 - Electrolyzer Voltage - 1.58 V; Temperature - 100°C
Case 2 - Electrolyzer Voltage - 1.68 V; Temperature - 100°C
Case 3 - Electrolyzer Voltage - 1.78 V; Temperature - 100°C
Case 4 - Electrolyzer Voltage - 1.88 V; Temperature - 60°C
Case 5 - Electrolyzer Voltage - 1.63 V; Temperature - 150°C

Figure 4-11. Sulfuric Acid Electrolyzer Overall Plant Efficiency (100°C)
Versus Cell Voltage

Figure 4-12. Sulfuric Acid Electrolyzer Overall Plant Efficiency (200 mA/cm^2) Versus Electrolyte Temperature

that there is little increase in the conductivity of sulfuric acid beyond the temperature of 100°C. Therefore, there is not any incentive, from the efficiency viewpoint, to operate the electrolyzer at a temperature much higher than 100°C.

SULFURIC ACID ELECTROLYSIS PLANT ECONOMIC EVALUATION

The economics for the sulfuric acid electrolysis plant were evaluated using the same cost assumptions as applied in the Sulfur Cycle analysis. The economic evaluation considered utility financing, 18 percent annual charge on depreciating investments, an 80 percent plant capacity factor, a 1976 cost basis, and no credit for the oxygen produced. The evaluation was made, as in the Sulfur Cycle, for a 380 million SCFD grass roots self-sufficient hydrogen production plant energized by a VHTR. The electrolyzer geometrical design and cell configuration were similar to that in the Sulfur Cycle, except for different electrode materials and catalysts.

Table 4-6 shows the estimate of direct capital cost for the hydrogen production plant. The costs are arranged in a format similar to the Sulfur Cycle Process, with the same designation of Batteries for cost accounting. The differences are that the Sulfur Cycle on-sites Batteries, H, Sulfuric Acid Decomposition, I, SO_2/O_2 separation, J, Turbine Generation System, and the off-sites chilled water system are not needed, since these Batteries apply only to the Sulfur Cycle and all electric power is now produced in the VHTR plant. The effect of the substantially larger electric power requirements of the sulfuric acid electrolyzers, as compared to the depolarized electrolyzers in the Sulfur Cycle plant, is quite apparent in the cost of Battery F, Electrolyzer Power Supply. The cost of Battery G, Electrolyzer and Auxiliaries, is shown in more detail in Table 4-7.

The direct capital costs for the hydrogen production facility are relatively constant, as shown in Figures 4-13 and 4-14. The differences are primarily due to increased power requirements as voltages increase. Overall, the total direct capital cost for the sulfuric acid hydrogen production facility is about \$100 million less than that calculated for the Sulfur Cycle. Because of the increased power required for the sulfuric acid electrolyzer, this decrease in the direct capital cost of the hydrogen production facility is compensated for by an increase in VHTR cost.

Table 4-6

SULFURIC ACID PLANT
DIRECT CAPITAL COST - 1976 BASIS
(Thousands of Dollars)

	<u>Case 1</u>		<u>Case 2</u>		<u>Case 3</u>		<u>Case 4</u>		<u>Case 5</u>	
<u>Electrolyzer Voltage Cases</u>	<u>1.58v</u> <u>100°C</u>		<u>1.68v</u> <u>100°C</u>		<u>1.78v</u> <u>100°C</u>		<u>1.88v</u> <u>60°C</u>		<u>1.63v</u> <u>150°C</u>	
<u>On-Sites</u>	<u>\$10³</u>	<u>\$/kW⁽²⁾</u>	<u>\$10³</u>	<u>\$/kW⁽²⁾</u>	<u>\$10³</u>	<u>\$/kW⁽²⁾</u>	<u>\$10³</u>	<u>\$/kW⁽²⁾</u>	<u>\$10³</u>	<u>\$/kW⁽²⁾</u>
Battery F - Electrolyzer Power Supply	\$ 96,500	\$ 64.0	\$ 97,492	\$ 64.6	\$ 98,485	\$ 65.3	\$ 99,469	\$ 66.0	\$ 96,998	\$ 64.3
Battery G - Electrolyzer & Aux.	238,714	158.3	239,044	158.5	239,245	158.6	238,678	158.3	239,002	158.5
Battery J - Turbine Generation System					(Included as part of the VHTR Plant Costs)					
On-Sites Sub-Total	\$335,214	\$222.3	\$336,536	\$223.1	\$337,730	\$223.9	\$338,147	\$224.3	\$336,000	\$222.8
<u>Off-Sites</u>										
Cooling Water System & Water Intake	\$ 9,678	\$ 6.4	\$ 10,342	\$ 6.9	\$ 10,935	\$ 7.3	\$ 10,518	\$ 7.0	\$ 10,092	\$ 6.7
Makeup & Feedwater	1,973	1.3	1,973	1.3	1,973	1.3	1,973	1.3	1,973	1.3
Waste Water Treatment	340	0.2	340	0.2	340	0.2	340	0.2	340	0.2
Electrical Aux. Power	11,045	7.3	11,950	7.9	12,759	8.5	11,672	7.7	11,616	7.7
Land & Land Rights ⁽¹⁾	200	0.1	200	0.1	200	0.1	200	0.1	200	0.1
Site Improvements & Facilities	1,080	0.7	1,080	0.7	1,080	0.7	1,080	0.7	1,080	0.7
Admin./Service/Lab Bldgs.	1,430	0.9	1,430	0.9	1,430	0.9	1,430	0.9	1,430	0.9
Instrument & Plant Air	2,650	1.8	2,650	1.8	2,650	1.8	2,650	1.8	2,650	1.8
Maintenance Facilities	1,550	1.0	1,550	1.0	1,550	1.0	1,550	1.0	1,550	1.0
Fire Protection, Communication	950	0.6	950	0.6	950	0.6	950	0.6	950	0.6
Furnishings, Fixtures, etc.	250	0.2	250	0.2	250	0.2	250	0.2	250	0.2
Off-Sites Sub-Total	\$ 31,146	\$ 20.5	\$ 32,715	\$ 21.6	\$ 34,117	\$ 22.6	\$ 32,613	\$ 21.5	\$ 32,131	\$ 21.2
Total Direct Capital Cost	\$366,360	\$242.8	\$369,251	\$244.7	\$371,847	\$246.5	\$370,760	\$245.8	\$368,131	\$244.0

(1) Some items in General Off-Site are shared with VHTR and included partially in VHTR costs.

(2) Based on Heating Value of Product Hydrogen.

Table 4-7

SULFURIC ACID PLANT
BATTERY G - ELECTROLYZER AND AUXILIARIES
DIRECT CAPITAL COST - 1976 BASIS

(Thousands of Dollars)

<u>Item</u>	<u>Case 2 (1.68 Volts)</u>
Electrolyzers	\$194,873
• Cells	\$112,464
• Vessel	\$ 35,616
• Internal Structure, etc.	\$ 46,793
Electrolyzer External Supports and Installation	2,300
Electrolyzer Instrumentation	3,700
Electrolyzer Auxiliary Equipment, Installed ⁽¹⁾	13,261
D-C Bus Duct, Installed	21,200
Electrolyzer Building	<u>3,710</u>
Total	\$239,044

⁽¹⁾ Includes installed cost of equipment closely associated with electrolyzer, including piping, valves, supports, instrumentation, etc., as shown in Figure 4-4.

Figure 4-13. Sulfuric Acid Electrolyzer Plant Direct Capital Cost Versus Cell Voltage - 1976 Basis (100°C)

Figure 4-14. Sulfuric Acid Electrolyzer Plant Direct Capital Cost Versus Electrolyte Temperature - 1976 Basis (200 mA/cm²)

The total plant investment is given in Table 4-8 and includes both the hydrogen production facility and the VHTR energy source. As in the Sulfur Cycle evaluation, the nondepreciating assets are land and land rights. The special materials comprise the initial supply of coolants, chemicals, lubricants, and other materials needed for operation of the overall plant. A 20 percent contingency is applied to the hydrogen production and VHTR plant direct costs to account for unknowns and estimating inaccuracies.

Indirect costs are expense items of a general nature which apply to the overall project of building an operable plant, rather than to one of the direct costs. These costs, except for interest during construction, have not been estimated in detail, but calculated as a percentage of the direct costs in the same manner as in the Sulfur Cycle evaluation. For all cases, they have been apportioned between the VHTR and the hydrogen production plant. Interest during construction is calculated as simple interest, at an 8 percent annual rate, for an eight year project schedule.

A summary plot of the sulfuric acid electrolyzer plant total capital cost as a function of cell voltage, at constant temperature, is shown in Figure 4-15. The hydrogen production and the VHTR plant contribution to the total investment is also shown. In a similar way, Figure 4-16 shows the total capital cost as a function of electrolyte temperature.

The cost of the sulfuric acid electrolysis plant will also vary with the current density employed. Figure 4-17 shows that variation. The curves assume that the voltage shown is achieved at 200 mA/cm^2 . As the current density increases, the voltage is assumed to increase. Therefore, as current density increases for a constant hydrogen output, the investment tends to decrease due to lower electrolyzer costs and increase due to greater electrolyzer power requirements. The net effect is a flattening of the investment curve as the current density approaches 600 mA/cm^2 .

The hydrogen production cost for each of the voltage and temperature cases is given in Table 4-9 and is made up of the contributions of capital, operation and maintenance, and nuclear fuel cost. These are calculated on an annual basis. As was done for the Sulfur Cycle process: 1) a 10 percent annual charge was assumed for land; 2) an 18 percent annual charge was used on depreciating assets;

Table 4-8

SULFURIC ACID PLANT
CAPITAL COST SUMMARY - 1976 BASIS

Electrolyzer Voltage Cases	(Thousands of Dollars)									
	Case 1		Case 2		Case 3		Case 4		Case 5	
	1.58v 100°C		1.68v 100°C		1.78v 100°C		1.88v 60°C		1.63v 150°C	
	VHTR	H ₂ Prod	VHTR	H ₂ Prod	VHTR	H ₂ Prod	VHTR	H ₂ Prod	VHTR	H ₂ Prod
<u>Non-Depreciating Assets</u>										
Land & Land Rights	\$ 800	\$ 200	\$ 800	\$ 200	\$ 800	\$ 200	\$ 800	\$ 200	\$ 800	\$ 200
<u>Depreciating Assets</u>										
Special Materials	350	370	350	370	350	370	350	370	350	370
Physical Plant	363,790	366,160	377,749	369,051	391,430	371,647	404,333	370,560	371,092	367,931
Sub-Total	364,140	366,530	378,099	369,421	391,780	372,017	404,683	370,930	371,442	368,301
Contingency (20%)	72,828	73,306	75,620	73,884	78,356	74,403	80,937	74,186	74,288	73,660
Sub-Total	436,968	439,836	453,719	443,305	470,136	446,420	485,620	445,116	445,730	441,961
<u>Indirect Costs</u>										
Construction Fac., Equip., Services	28,228	9,237	29,310	9,309	30,371	9,375	31,371	9,347	28,794	9,281
Engineering Serv.	68,604	15,394	71,234	15,516	73,811	15,625	76,242	15,579	69,980	15,469
Other Costs	21,761	3,959	22,595	3,990	23,413	4,018	24,184	4,006	22,197	3,978
Interest During ⁽¹⁾ Constr. (8% Yr.)	<u>151,541</u>	<u>75,652</u>	<u>157,350</u>	<u>76,248</u>	<u>163,043</u>	<u>76,784</u>	<u>168,413</u>	<u>76,560</u>	<u>154,579</u>	<u>76,017</u>
Total Depreciating Assets	707,102	544,078	734,208	548,368	760,774	552,222	785,830	550,608	721,280	546,706
Total Plant Investment	\$707,902	\$544,278	\$735,008	\$548,568	\$761,574	\$552,422	\$786,630	\$550,808	\$722,080	\$546,906
	<u>\$1,252,180</u>		<u>\$1,283,576</u>		<u>\$1,313,996</u>		<u>\$1,337,438</u>		<u>\$1,268,986</u>	
	(\$830/kW) ⁽²⁾		(\$851/kW) ⁽²⁾		(\$871/kW) ⁽²⁾		(\$887/kW) ⁽²⁾		(\$841/kW) ⁽²⁾	

(1) Eight-Year Construction Schedule

(2) Based on Heating Value of Product Hydrogen

Figure 4-15. Sulfuric Acid Electrolyzer Plant Capital Cost Versus Cell Voltage - 1976 Basis (100°C)

Figure 4-16. Sulfuric Acid Electrolyzer Plant Capital Cost Versus Electrolyte Temperature - 1976 Basis (200 mA/cm^2)

Figure 4-17. Sulfuric Acid Electrolysis Total Plant Investment Versus Current Density

Table 4-9

SULFURIC ACID PLANT
HYDROGEN PRODUCTION COST (NO OXYGEN CREDIT)

80% Capacity Factor
Utility Financing
1976 Basis

<u>Electrolyzer Voltage Cases</u>	<u>Case 1</u>	<u>Case 2</u>	<u>Case 3</u>	<u>Case 4</u>	<u>Case 5</u>
	1.58v 100°C	1.68v 100°C	1.78v 100°C	1.88v 60°C	1.63v 150°C
<u>Annual Costs (Thousands of Dollars)</u>					
Non-Depreciating Capital ⁽¹⁾	\$ 100	\$ 100	\$ 100	\$ 100	\$ 100
Depreciating Capital ⁽²⁾	225,212	230,864	236,339	240,559	228,238
Operation & Maintenance	6,394	6,670	6,940	7,196	6,538
Nuclear Fuel Cycle Cost ⁽³⁾	<u>41,546</u>	<u>44,236</u>	<u>46,939</u>	<u>49,546</u>	<u>42,945</u>
Total Annual Cost	\$273,252	\$281,870	\$290,318	\$297,401	\$277,821
 Annual H ₂ Production (SCF)					
	1.11 x 10 ¹¹	1.11 x 10 ¹¹	1.11 x 10 ¹¹	1.11 x 10 ¹¹	1.11 x 10 ¹¹
 <u>Hydrogen "Gate Price"</u>					
\$ / MSCF	2.46	2.54	2.62	2.68	2.50
\$ / 10 ⁶ BTU	7.56	7.80	8.05	8.23	7.68

(1) Land at 10% Annual Charge

(2) 18% Annual Charge

(3) At 50¢/10⁶ BTU for Nuclear
Fuel Cost

3) an 80 percent capacity factor was assumed for utility financing; and 4) a $50\text{¢}/10^6$ Btu nuclear fuel cost was used. The data in the table is plotted in Figure 4-18 to show the hydrogen production "gate price" as a function of cell voltage.

To account for the variability of hydrogen production costs with nuclear fuel cost, Figure 4-19 shows hydrogen "gate price" as a function of fuel cost for the various voltage and temperature cases. Doubling the nuclear fuel cost results in a 15 percent increase in hydrogen "gate price." This is somewhat higher than the effect in the Sulfur Cycle plant and results from the greater relative contribution of fuel cost to the production cost of the lower efficiency sulfuric acid electrolysis plant.

The effect of current density on hydrogen "gate price" is shown in Figure 4-20 for nuclear fuel costs of $\$0.50/10^6$ Btu, $\$1.00/10^6$ Btu, and $\$1.50/10^6$ Btu, respectively. As in the Sulfur Cycle study, these cost curves tend to flatten as the current density is increased.

Figure 4-18. Sulfuric Acid Plant Hydrogen "Gate Price" Versus Cell Voltage - 1976 Basis (50¢/10⁶ Btu Nuclear Fuel Cost)

Figure 4-19. Sulfuric Acid - Electrolyzer Plant Hydrogen
"Gate Price" Versus Nuclear Fuel Cost - 1976 Basis
1976 Basis

Figure 4-20. Sulfuric Acid Electrolysis Versus Current Density
(Nominal 1.68 Volt at 200 mA/cm²)

Section 5

COMPARATIVE HYDROGEN PRODUCTION EFFICIENCY AND ECONOMICS

The evaluation of the two hydrogen production systems has shown that the Sulfur Cycle efficiency is about six percentage points higher than the efficiency of the sulfuric acid electrolysis plant, when both processes are operating at cell voltages believed to be reasonably attainable; i.e., 600 mV for the Sulfur Cycle and 1.68 volts for the sulfuric acid electrolysis. Figure 5-1 shows how the plant efficiencies vary with cell voltage. The figure can also be used to determine the cell voltages needed in both systems in order to have equal efficiencies. For example, with the Sulfur Cycle at 600 mV (an overall efficiency of 47 percent) the sulfuric acid electrolyzer would have to operate at about 1.43 volts in order to achieve a parity in efficiency. This is below the thermal neutral voltage for water electrolysis and, therefore, may not be a practical operating point even if achievable.

The economic comparison of the cost of hydrogen produced from the Sulfur Cycle process and Sulfuric Acid Electrolysis process indicates that, for cell voltages of 600 mV and 1.68 volts, respectively, the Sulfur Cycle hydrogen production costs are lower. With the economic model adopted for this study, hydrogen can be produced by the Sulfur Cycle for approximately \$2.38/MSCF. This cost is almost 16¢/MSCF lower than the Sulfuric Acid Electrolyzer production cost. Figure 5-2 shows the hydrogen "gate price" versus cell voltage. For economic parity with the Sulfur Cycle operating at a cell voltage of 0.6 volts, the Sulfuric Acid Electrolyzer would have to operate at approximately 1.48 volts.

A comparison of the sulfur cycle plant and sulfuric acid electrolysis plant hydrogen "gate price" is shown in Figure 5-3 for the nominal voltage cases of 0.6 volts and 1.68 volts, respectively. Here, hydrogen production costs are plotted against nuclear fuel cost in $\text{¢}/10^6 \text{ BTU}$. The Sulfur Cycle hydrogen production costs are lower by 15¢/MSCF to 30¢/MSCF than costs from the sulfuric acid electrolysis over the range of nuclear fuel costs considered. For both processes, doubling the nuclear fuel cost results in only a 13 to 15 percent increase in hydrogen "gate price."

Figure 5-1. Comparison of Overall Plant Efficiencies Versus Cell Voltage

Figure 5-2. Comparison of Hydrogen "Gate Price" Versus Cell Voltage - 1976 Basis
(\$0.50/10⁶ Btu Nuclear Fuel Cost)

Figure 5-3. Sulfur Cycle Plant Comparison with Sulfuric Acid Electrolyzer Plant Hydrogen "Gate Price" Versus Nuclear Fuel Cost

Section 6

CONCLUSIONS

In comparing the performance and economics of the two hydrogen production systems, it must be borne in mind that both systems, and the nuclear energy source for both, require further development to substantiate the engineering judgements used in defining flow sheets, performance capabilities, component designs, and economics. The overall conclusions of the study using a consistent design and costing approach for both systems, are:

- There is a consistent difference in overall thermal efficiency of hydrogen production for the two concepts, with the Sulfur Cycle having an efficiency of some six percentage points higher (47 percent versus 40.9 percent) than the sulfuric acid electrolysis plant, when both are evaluated on the basis of reasonably expected electrolysis performance levels.
- The economics, recognizing the caveats of cost estimates on systems and components not yet fully developed, shows the hydrogen production cost of the Sulfur Cycle plant to be approximately six to seven percent lower than that of the sulfuric acid electrolysis plant. Because of the lower efficiency of the latter, the effect of increasing fuel costs is to increase the production cost difference between the two systems.
- The development requirements for the nuclear energy source are essentially the same regardless of which hydrogen production system is employed. While not considered during this study, a sulfuric acid electrolysis system could, of course, be driven by existing methods of power generation that require no development.
- The electrolysis step is a critical element of both hydrogen production systems. The cost and efficiency of the electrolysis step is, in both systems, a major determinant of hydrogen cost and overall efficiency.
- The development program required to commercialize the hydrogen production systems themselves are substantially different. The sulfuric acid electrolysis system will require much less funding and time than the Sulfur Cycle program to achieve its objectives. This is because it not only benefits from the technological fallout from other electrolysis programs, but it does not have to contend with the high temperature process steps and materials for high temperature applications required in the Sulfur Cycle.

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APPENDIX A

VHTR HEAT SOURCE

BACKGROUND

The VHTR is a thermal reactor that uses helium as coolant and has an all-ceramic core composed of a graphite moderator and enriched uranium-thorium fuel. This combination has enabled the gas-cooled reactor to develop coolant outlet temperatures far higher than those of other reactor systems. For production of electricity, these temperatures permit the production of high-temperature, high-pressure steam, leading to high-cycle efficiency. Similar designs, at more elevated temperatures, can be useful for generating process heat.

In 1974, the U.S. Atomic Energy Commission (now DOE) embarked on a program to assess and initiate the development of a class of very high temperature nuclear reactors, VHTR, which would be uniquely capable of meeting a segment of the energy market requiring process heat temperatures beyond the capability of other nuclear heat sources. The VHTR technology was to be based on the AEC/NASA nuclear rocket engine program (NERVA), the helium cooled gas cooled reactor (HTGR) being developed at that time for the generation of steam for electric utility plants, and the German program for the development of gas cooled pebble bed reactors.

In late 1974, conceptual design studies (1,2,3) were published by the General Atomic Company, General Electric Company, and the Westinghouse Advanced Energy Systems Division (formerly the Astronuclear Laboratory). These studies reflected three different approaches to the design of a VHTR, i.e., a slightly modified HTGR (GA), which, with minimum development, could modestly increase reactor outlet coolant temperature; a pebble bed reactor (GE), patterned after the German AVR reactor, which by virtue of a directly cooled low power density core could achieve desirably high reactor temperatures; and a prismatic core gas-cooled reactor (Westinghouse), using fuel elements derived from the nuclear rocket program, that would be limited in temperature capability only by the availability of high temperature structural materials required to transfer the heat to the process.

Table A-1 summarizes the major features of the three VHTR designs prepared by GA, Westinghouse and GE. Each of the reactor manufacturers also prepared, during their 1974 studies, the design of a "nuclear island" employing features each judged appropriate to the process needs and most adaptable to the nuclear system. In addition, capital, fuel, and operation costs for the plant were estimated, along with development requirements and schedules for commercialization of the concept.

The need for the VHTR, as shown by studies done in the U.S. and the programs underway elsewhere in the world, e.g., Germany and Japan, gives impetus to the establishment of a well defined development program that addresses the remaining technical issues and provides demonstration of the nuclear system.

DESCRIPTION OF VHTR DESIGN APPROACHES

General Atomic developed the HTGR as a commercial steam-generating reactor for use by utilities. The Peach Bottom Reactor [115 MW(t), 40 MW(e)] was operated successfully by Philadelphia Electric Company from June 1967 to October 1974 and proved the feasibility of the HTGR.

General Atomic has built a much larger HTGR at the Fort St. Vrain Nuclear Generating Station [842 MW(t), 330 MW(e)] which is currently in its start-up program leading to full power operation. The standard HTGR (helium outlet temperature of 1400°F) was modified for process heat purposes by placing a process heat exchanger in series with and upstream of the steam generator. As in other HTGR designs, all the primary system components are contained in one large prestressed concrete reactor vessel (PCRV) because of the advantages offered by this form of construction. Steam-methane reforming was selected by GA as a typical high-temperature process; the reformer was assumed to be located within the PCRV. The GA study was based on the assumption that an intermediate heat exchanger (IHX) would not be required. If an IHX is required, reactor exit helium temperatures would have to be increased well above the reference 1620°F to reach their postulated 1400°F maximum process temperature.

The Westinghouse VHTR contains a number of attractive features. The reactor and its coolant loops are contained within a multicavity prestressed cast iron reactor vessel (PCIV). The reactor core is located within the vessel's central cavity. The vessel walls contain 12 smaller vertical cavities, or pods, in which are five high-temperature intermediate heat exchangers and circulators, five gas turbines and low-temperature intermediate heat exchangers, and two auxiliary cooling systems for shutdown and emergency cooling of the reactor. Reactor helium coolant

Table A-1
COMPARISON OF VHTR DESIGNS
(Reference 4)

Table 8. Comparison of the three vendor concepts

	General Atomic	General Electric	Westinghouse
Basis of very high-temperature nuclear reactor design	Modification of HTGR concept	Pebble bed concept based on German technology	Prismatic fuel concept based on nuclear rocket technology
Reactor core type	Hexagonal graphite blocks containing separate coolant and fuel passages, solid cylindrical fuel rods	Pebble bed core Graphite sphere fuel element	Hexagonal graphite blocks, hollow cylindrical fuel rods with central coolant channel
Reactor coolant	Helium	Helium	Helium
Reference thermal power	3000 MW(t)	3000 MW(t)	3000 MW(t)
Pressure vessel concept	Prestressed concrete reactor vessel	Prestressed concrete reactor vessel	Prestressed cast iron reactor vessel
Fuel composition	Fully enriched U feed (UC ₂) with thorium (ThO ₂) fertile material TRISO coating for both fissile and fertile particles	Low enriched (9.01%) ²³⁵ U fuel Uranium-thorium fuel alternate TRISO-coated UO ₂ fuel particles	Fully enriched U feed (UC ₂) with thorium (ThO ₂) fertile material TRISO-coated fissile particles BISO-coated fertile particles
Average fuel residence time	3 years	3.8 years	4 years
Power density of core	8.4 W/cm ³	5 W/cm ³	10 W/cm ³
Reactor core ΔP	16 psi	6 psi	5 psi
System pressure	725 psi	600 psi	1000 psi
Core inlet temperature	770°F	482°F	807°F
Reference maximum process temperature	1400°F (without IHX)	1500°F (with IHX)	1600°F (with IHX)
Core outlet temperature	1600°F	1742°F	1850°F
Maximum fuel temperature	2475°F	2030°F	2158°F
Type process	Steam-hydrocarbon reformer	Steam-hydrocarbon reformer	Hybrid thermochemical-electrolytic water splitting

enters and discharges from the pods through coaxial piping at the upper end of the cavity, while the intermediate loop, or secondary, helium coolant is introduced and leaves through the bottom of the pod. The PCIV has a continuous internal steel liner to act as a primary coolant boundary and leak-tight membrane. A thermal barrier and insulation system is used to limit the temperature of the liner and minimize the heat loss to the PCIV.

The process heat exchangers and the gas turbine-generators provide energy for the Westinghouse hybrid process for hydrogen generation.

The pebble-bed reactor (PBR) developed by Kernforschungsanlage (KFA), Julich, differs from the HTGR primarily by having the fuel contained in graphite moderator balls. These balls are loaded into and withdrawn from the reactor during operation. The PBR concept was selected by GE for their VHTR design.

KFA has built and is operating the Arbeitsgemeinschaft Versuchsreaktor (AVR) [46 MW(t), 15 MW(e)], an experimental steam-generating PBR. The AVR has operated at a helium core outlet temperature of 1742°F since February 1974 (5). A larger PBR, the high-temperature reactor (THTR) [768 MW(t), 300 MW(e)], is under construction (6) and is scheduled for 1980 (7) operation. This steam-generating reactor will have a helium outlet temperature of 1380°F, in the same range as do commercial HTGRs.

The GE PBR places all primary system components into a PCRV. Primary helium coolant flows through the core and exists into five loops, each containing an IHX and a steam generator. A secondary helium stream is heated in the IHX and is transported out of the reactor vessel to an external process heater, which GE assumes to be a methane-steam reformer.

All three reactor concepts appeared to have potential to achieve process temperatures in the range 1200 to 2000°F. The GE concept has the most applicable operational background in that the AVR has been operated successfully at 950°C for extended periods. General Atomic, on the other hand, has developed technology applicable to 3000-MW(t) designs and currently has the broadest technological base. The Peach Bottom reactor outlet temperature was only 1320°F; however, it appears likely that the HTGR technology, with development, would be applicable to more elevated temperatures. The Westinghouse concept does not have a

commercial technological base comparable to the other two concepts; it is based however on the fuel element technology successfully demonstrated in the nuclear rocket program at coolant temperatures up to 4000°F.

The process temperature of each of the reactor concepts of Table A-1 is indicative of the historical development of the concepts. The HTGR has evolved from a technology program focused on steam generation. The lowest temperature required for a reasonable process efficiency, 1400°F was chosen to minimize extrapolation of existing designs and technology. The PBR concept is based on the technology of the AVR scaled up to commercial size; the process temperature selection of 1500°F is a logical consequence of the AVR helium outlet temperature of 1740°F assuming that the heat transport systems can be developed. The Westinghouse concept is derived from the very advanced NERVA technology, which operated at extremely high temperatures. The water decomposition process driven by the Westinghouse VHTR was based on a 1600°F peak process temperature.

THE VHTR DEVELOPMENT PROGRAM

The VHTR development program is a multi-national program aimed at achieving a demonstration of a helium cooled reactor operating with outlet temperatures of about 1000°C, suitable for use in a "process" configuration. The lead countries in initiating the VHTR program were the USA, Federal Republic of Germany, and Japan. The program plans for these three countries were first presented at the IAEA Symposium on "Gas Cooled Reactors with Emphasis on Advanced Systems," held in Julich, FRG, in October, 1975 (8,9,10).

Since the time of the Symposium, the character of the multi-national program has evolved. The German program, based on the pebble bed reactor, has proceeded with the continued operation of the AVR, construction of the THTR, and experimental work in materials, components, and techniques to permit a demonstration plant to be designed and ultimately built.

The Japanese program, although smaller than the German program, has also made strides in the intervening years. The need for the VHTR continues to exist and receive governmental support (11). Test facilities and experimental programs are being diligently pursued (12,13). The plans call for an experimental VHTR, rated at 50 MW(t) and operating at 1000°C, to be operational by 1984 (14).

The planned program in the U. S. has been severely cut back from the plans laid out in 1975 (10). The present U. S. approach is to operate in a cooperative manner with the FRG, under a ten year agreement entered into in February, 1977, to jointly develop the technology of the VHTR. The U. S. funded contribution to the program now consists of an effort to assess the compatibility of structural materials with high temperature helium and a number of studies whose purpose it is to compare the features, economics, and licensing characteristics of the German pebble bed and the U. S. "prismatic" core VHTRs for the purpose of defining the most appropriate VHTR type for U. S. applications. Cooperative programs in fuels, materials, components, and demonstrations are being discussed between the two countries; initiation of these programs remains to be accomplished.

The assessment which can be made of the VHTR development program is that those countries which clearly have seen a national need for the reactor, i.e., Germany and Japan, are aggressively pursuing the program. The U. S., on the other hand, has a less imperative need (e.g., large resources of coal to provide high temperature process heat) and therefore a lower priority and funding level for the VHTR. The VHTR is, nevertheless, under development and should be available as a commercial heat source, albeit imported from overseas, around the turn of the century.

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APPENDIX B

DEVELOPMENT REQUIREMENTS

The development requirements for the two hydrogen production processes considered in this study are similar in some respects and quite dissimilar in others. There is a commonality in the electrolyzers, with the development areas being the same, although the problems to be solved are somewhat different. The most striking departure between the two development programs is, of course, the need to develop high temperature materials, catalysts, and components for the thermal steps of the hybrid Sulfur Cycle Water Decomposition System.

SULFUR CYCLE WATER DECOMPOSITION

The program for the development of the Sulfur Cycle has been under way since 1973, with Westinghouse funding, and has been supported by the Department of Energy since 1976. The development program plan, as currently conceived, is a multi-year program leading to a pre-pilot integrated bench scale cyclic process demonstration unit (PDU) by 1983. Pilot scale and/or demonstration units would follow after the PDU, but are not specifically identified in the present plan.

Figure B-1 indicates the key items in the program and their schedular projections. There are four major areas to be treated in the overall program. These are the electrolyzer (Task 1.0), Sulfur Trioxide Reduction (Task 2.0), Process Studies (Task 3.0), and Integrated Testing (Task 4.0). The sections below describe the effort to be accomplished in each area for the duration of the program.

Task 1.0 Electrolyzer

The SO_3 depolarized electrolyzer is the component in which hydrogen is produced. The development program is organized to consider the electro-catalysts, electrodes, cell membranes, pressurized and bi-polar cell design and performance, and scale-up of test articles to demonstrate the potential of meeting performance and economic objectives.

Figure B-1. Sulfur Cycle Development Program

As shown in Reference 1, the anode reaction taking place in the cell maybe written as :

while the cathode reaction is

The reactions could result in electrode potentials as shown in Figure B-2. The overall cell potential, less overpotentials and IR losses, would be the difference between the anode and cathode curves shown in the figure.

Early in the Sulfur Cycle program, it was believed that anode reaction (a) would best describe the performance capability of the cell. Based on that belief, the cell design for the conceptual design prepared for NASA in 1975 (2), which used a 75 w/o sulfuric acid electrolyte, was 450 mV (255 mV cell potential plus an allowance of 195 mV for overpotentials and IR losses). Similarly, the subsequent flow sheet for the process, prepared in late 1976 and reported in Reference 1, was based on an 80 w/o acid electrolyte, a cell potential of 275 mV, an allowance of 205 mV for overpotentials and IR losses, and a resulting total cell voltage of 480 mV.

The results of more recent experimental work, in conjunction with further analytical evaluation, gives indications that the assumption that only anode reaction (a) occurs may be overly optimistic. The present view is that the anode reaction may be some combination of reactions (a) and (b), but no worse, in terms of electrode potential, than reaction (b). The determination of the actual conditions in the cell must ultimately be determined experimentally. In the interim, one can project total cell voltages only over a range reflecting the uncertainties. For example, assuming that a total 200 mV loss for overpotentials and IR is achievable, the total cell voltage may be 400 to 485 mV at 50 w/o acid concentration, 435 to 555 mV at 60 w/o acid concentration, and 475 to 720 mV at 80 w/o acid concentration.

Figure B-2. The Effect of Acid Concentration on Electrode Potential
(Calculated for Different Ionizations)

The experimental program in electrocatalysts, electrodes, and pressurized cells is designed to determine the electrode potentials, overpotentials, and IR losses that can be achieved in the progression towards a "commercial" cell.

The cell overpotentials are considered in several subtasks. The electrocatalyst task is searching for better and, hopefully, less expensive catalyst materials for both the anode and cathode. The program in electrodes is concerned with both anodes and cathodes. The pressurized cell will permit higher concentrations of SO_2 in the anolyte (even at higher operating temperatures) than has been achieved to date, increasing the ability to effectively use the anode material.

Improvements in IR losses will result from higher operating temperatures (lower acid resistivity) to be demonstrated in the pressurized cell plus the improvements in membranes (thinner, stronger, lower resistivity) expected.

The technology developed in each of the tasks will be incorporated into the laboratory model bi-polar cell (a stack of five or six cells with an electrode area of about 25 square inches operating at atmospheric pressure), the scale-up cell (a bi-polar stack of five or six cells with an electrode area of about one square foot operating at 300 psi), and the PDU cell (size yet to be determined).

Task 2.0 Sulfur Trioxide Reduction

The task called Sulfur Trioxide Reduction covers that part of the hydrogen generation process that takes sulfuric acid from the electrolyzer, concentrates and vaporizes the stream, and reduces the resulting steam/ SO_3 mixture to steam, SO_2 , O_2 , and unreacted SO_3 . The scope of this task includes not only the catalyst investigations for the reduction reaction itself, but also the materials of construction for the high temperature acid vaporizer and reduction reactors. The applicability of this part of the process to other end uses, such as the use of the endothermic reduction reaction as a means to store energy, requires that sufficient flexibility be designed into the experimental program to assure data applicable to a spectrum of applications.

The continuing capability to quickly and inexpensively screen candidate reduction catalysts to be able to determine those with high potential for economically attractive activities and life times is an important aspect of demonstrating the merit of the process. Accordingly, an atmospheric test facility is employed to

assess the activities and aging characteristics of selected catalysts over the temperature range of interest. The process fluid to be reduced will contain SO_3 and water vapor in varying proportions to simulate the inlet conditions for a range of sulfuric acid concentrations.

While the catalysts screening is done at atmospheric pressure, commercial reactors would operate at pressures up to perhaps 300 psi. The effect of pressure on the decomposition kinetics has to be explored. Therefore, a pressurized catalytic sulfur trioxide reduction reactor is an important development tool.

Using the pressurized reactor, the kinetics of sulfur trioxide decomposition will be examined for space velocities between 5000 and 60,000 hr^{-1} , temperatures between 800 and 1600°F, and for pressures to 300 psi. Inlet compositions will correspond to the sulfur trioxide mole fraction, in steam, obtained by vaporizing several different acid concentrations. Not all catalysts examined in the catalyst screening program would be tested in the pressurized system. Only the more promising candidates would be subjected to the more extensive test program.

The compatibility of containment materials with the process stream at each step where heat is added or removed is an important consideration affecting the feasibility of the water splitting process. There are little or no data available with regard to the compatibility of structural materials with sulfuric acid under the conditions indicated for the acid vaporization step. The vaporizer materials are aimed at resolving material uncertainties and uncovering, at an early stage, material problems which may impact the process.

The high temperature SO_3 reduction reactor presents an additional challenge in respect to materials of construction. In order to confidently proceed to demonstrations of the process, suitable materials for operation in the environment of the reduction reactor must be identified and tested. Test beds will be operated to evaluate and screen candidate reduction reactor structural materials in a gas stream simulating the reactor inlet and outlet conditions. In addition, tests will be conducted on promising reduction catalysts to assure that they do not adversely interact with the reactor materials.

In the scale-up task, techniques and materials evaluated for the acid vaporizer materials and the reduction reactor materials test programs will be used to

design, construct, and operate a pilot loop which simulates the thermal acid decomposition and SO_2 reduction system. Operation of this loop will demonstrate materials feasibility of the process, as well as demonstrating the operation of the subsystem with parameters, materials, and flow streams typical of those expected in a large scale unit.

Task 3.0 Process Studies

Continued effort will be required in the performance of application studies, engineering trade-off studies, revision of process flow sheets, and economic analyses. The effect on the process flow sheet and performance of alternate heat sources, such as solar energy, will be evaluated. The integration of the process with the heat source will be evaluated. Also to be studied is the integration of the process with the end use application, be it hydrogen per se or a synthetic fuel, fertilizer, or the like. Results of the experimental program will be evaluated in respect to their effect on process performance and cost, and revisions made to the program as deemed appropriate by the evaluated effect of the data.

Task 4.0 Integrated Performance Testing

The purpose of this task is to provide feasibility and performance testing of the process at such times as the technology permits.

The laboratory model is an atmospheric pressure working model of the process. Such a model will provide early demonstration of the cyclic operation of the process, would provide the capability to assess interactions of the various process steps, and would provide an operating facility that can support the technical feasibility of hybrid electrochemical-thermochemical hydrogen production processes.

The purpose of bench scale PDU task may be simply stated—to demonstrate, with an integrated bench scale PDU, closed cycle operation of the process at operating conditions close to that desired for a commercial unit. The demonstration, as currently conceived, will include a multicell pressurized SO_2 depolarized electrolyzer which converts sulfurous acid and water into hydrogen and sulfuric acid; an acid vaporizer for converting aqueous sulfuric acid into steam and sulfur trioxide and a thermal reduction reaction for catalytically reducing sulfur trioxide

into sulfur dioxide and oxygen; and a recovery system for recycling water, sulfur dioxide, and unreacted sulfuric acid to the electrolyzer. This equipment will be capable of operating continuously for hundreds of hours in a fully closed cycle.

The equipment, catalysts, and materials employed in the PDU will, to the greatest extent possible, be the same as those which would be used in commercial systems. The electrolyzer will use those anodes, cathodes, and membranes deemed most appropriate under the Task 1.0 effort. The most promising reduction catalyst evaluated in Task 2.0 will be used for conducting the sulfur trioxide reduction reaction. The materials of construction of both the acid vaporizer and reduction reactor will be selected based upon the results of substantial materials evaluation programs.

With the backup of the effort on the electrolyzer and sulfuric acid decomposition programs, plus the process studies to identify and evaluate alternates, it is judged that the integrated bench scale PDU will confirm not only the capability of the process operating in a cyclic fashion, but also the efficiency levels and engineering approaches of a large scale commercial unit.

SULFURIC ACID ELECTROLYSIS

There is no current program devoted to the development of a water electrolyzer using sulfuric acid as the electrolyte. Development work on alkaline electrolyzers, solid polymer electrolyte (SPE) electrolyzers, and the SO₂-depolarized electrolyzer for the Sulfur Cycle can all contribute technology to a sulfuric acid electrolyzer and minimize the time and cost for a program.

The development program required for the sulfuric acid electrolyzer is completely analogous to the electrolyzer portion of the program shown in Figure 6-1 for the Sulfur Cycle process.

Work should be performed on electrocatalysts. The Sulfur Cycle electrocatalyst program as it pertains to the cathode is directly applicable; the presence of oxygen at the anode of the sulfuric acid electrolyzer indicates the need for an additional experimental effort to determine economically attractive catalytic materials that perform well in the presence of sulfuric acid and oxygen.

As in the case of the electrocatalysts, the electrode program on cathodes carried out for the Sulfur Cycle should be of direct value to the sulfuric acid electrolyzer. The carbon materials planned for the anodes of the Sulfur Cycle electrolyzer cannot, however, be used in the sulfuric acid electrolyzer because of the presence of oxygen at the anode. Development work is required, therefore, for the anode.

Most of the membrane development for the Sulfur Cycle is of value to the sulfuric acid electrolysis program. The membrane problem may be somewhat simpler for the sulfuric acid electrolyzer because there is no need for the permeability characteristics required to assure a flushing flow from the catholyte to the anolyte as in the Sulfur Cycle electrolyzer.

As in the Sulfur Cycle program, it is prudent to include a progression of test cells through pressurized cells and cell stacks up to prototypes of the commercial cell.

The "fallout" from other electrolysis programs leads to the judgment that a program for sulfuric acid electrolysis development can be defined that would lead to a large scale demonstration unit by 1982.

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