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CONFIGURING THE THERMOCHEMICAL HYDROGEN SULFURIC ACID PROCESS STEP FOR THE TANDEM MIRROR REACTOR

Terry R. Galloway

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Configuring the Thermochemical Hydrogen Sulfuric Acid Process Step for the Tandem Mirror Reactor

Terry R. Galloway
Lawrence Livermore National Laboratory
Livermore, California 94550

ABSTRACT

In joint program* between University of Washington, the General Atomic Company and ourselves, *we are developing a conceptual "point design" for a Tandem Mirror Reactor-driven thermochemical hydrogen plant.*

This paper identifies the sulfuric acid step as the critical part of the thermochemical cycle in dictating the thermal demands and temperature requirements of the heat source. We couple the General Atomic Sulfur-Iodine Cycle to a Tandem Mirror Reactor.

We focus specifically on the sulfuric acid decomposition process step since this step can use the high efficiency electrical power of the direct converter together with other thermal-produced electricity to Joule-heat a non-catalytic SO_3 decomposer to approximately 1250 K. This approach uses concepts originally suggested by Dick Werner and Oscar Krikorian. The blanket temperature can be lowered to about 900 K, greatly alleviating materials problems, the level of technology required, safety problems, and costs.

We have used a moderate degree of heat integration to keep the cycle efficiency around 48%, but limited the number of heat exchangers in order to keep hydrogen production costs within reasonable bounds.

INTRODUCTION

A projection of the energy supply and demand picture beyond the turn of the century shows (1) a substantial role for fusion both in the generation and in the production of synthetic fuels--starting with hydrogen.

Hydrogen, already a valuable raw material for our large U.S. petroleum and petrochemical industries, by early 2000 is expected to become an important renewable-based transportable fuel by itself or in some hydrocarbon form, such as methanol. Hydrogen can be produced through decomposition of water by means of thermochemical cycles, which reduce the high temperature 3000 K requirements of the straight thermal decomposition process to thermal levels that can be generated in nuclear fission or fusion reactors, or in high-intensity, focused solar collectors.

We have been studying the interfacing of our Tandem Mirror Fusion Reactor (TMR) to several thermochemical cycles under a DOE contract at LLL entitled: "Synfuels from Fusion" (2). *In this program we take advantage of the favorable engineering features of the TMR configuration.* The TMR consists of a cylindrical solenoidal, energy-producing section between end cells that provide the containment; the solenoid contains modular blanket sections with simple piping and electrical connections. The blanket produces heat. The direct converter produces both electricity and heat as the result of ion leakage out of the plasma into the end cells. The required tritium to fuel the DT reactor is also generated in the blanket modules. Materials and engineering structural problems are minimized by the [illegible] geometry, the steady state magnetic field and the temperature.

The chemical engineering portion of our program plan on this project is to develop a process design which is simple and credible, to keep the overall temperature requirement as low as possible, and capitalize on the unique advantages that fusion reactors offer as drivers to thermochemical hydrogen and [illegible] plant.

In his portion of the study, Dick Werner [illegible] the approximately 900 K cauldron [illegible], which [illegible] liquid metal such as [illegible] a large number of small modules which [illegible] the cylindrical plasma, and heat pipes [illegible] to transfer the heat from the liquid metal out into the [illegible].

The higher grade energy needed for the [illegible] temperature SO_3 decomposition process step at [illegible] K is supplied not from the blanket thermal [illegible] but, as initially proposed by R. Werner [illegible] high conversion efficiency electricity available in small quantities from the TMR [illegible] to electrically heat the SO_3 decomposer. [illegible] (3) we developed the conceptual design for a fluidized bed SO_3 decomposer (3), the heat for which was [illegible] *entirely from the blanket*; thus, [illegible] blanket temperature up to 1200 K. [illegible] Joule heated electrical elements [illegible] the blanket temperature [illegible] K.

Sulfur-Iodine Cycle Energy Requirement

The sulfur-iodine cycle being developed [illegible] the General Atomic Company (4) [illegible] thermochemical cycle operating [illegible]. The essential steps of the GA [illegible]

$$2H_2O + SO_2 + xI_2 \rightarrow H_2SO_4 + 2HI_x \quad [illegible] \quad (1)$$

$$2HI_x \rightarrow H_2 + xI_2 \quad [illegible] \quad (2)$$

$$H_2SO_4 \rightarrow H_2O + SO_2 + \tfrac{1}{2}O_2 \quad 1144\ K \quad (3)$$

All reactions in this system have been verified in the laboratory and total recycle has been demonstrated in a closed loop cycle, small-scale demonstrator. None of these reactions goes to completion; therefore, it is necessary to separate reaction products from the reactants. *Major parts of the process are associated with* separation and purification of the reaction products. A key to the successful operation of the process is the liquid-liquid phase separation of the reaction products of reaction (1). These solutions are separated into a lower density phase, containing H_2SO_4 and H_2O, and a higher density phase containing HI, I_2, and H_2O. This important advancement was developed at General Atomic.

Figure 1 is a schematic flow diagram of the sulfur-iodine cycle showing product mass flow and the recycle streams. The features shown in the conceptual block diagram in Fig. 1, are adequate for this paper.

In general, flowsheets of Section I involve H_2SO_4 and HI Production and Separation; Section II, H_2SO_4

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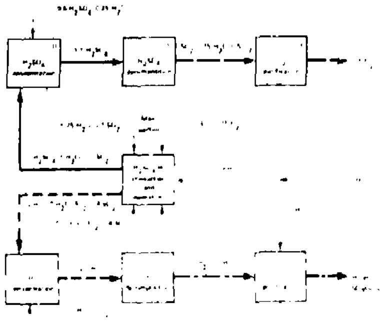

Fig. 1. Simplified schematic flow diagram of sulfur-iodine [illegible] (courtesy General Atomic Company).

Concentration and [illegible] Decomposition; Section III, HI Separation; Section IV, HI Decomposition and H_2 Purification [illegible]. The [illegible] reactor heat load for the [illegible] 60% [illegible] H_2.

The TMR [illegible] Interface

We [illegible] between the Tandem Mirror Reactor and the thermochemical H_2 cycle in Fig. 2. Heat [illegible] helium to make the process steam that [illegible] temperature process steps [illegible] over 14% of the TMR energy output [illegible] heat and [illegible] electricity from the [illegible].

We have [illegible] examined the engineering and safety aspects [illegible] the cycle by means of the more [illegible] liquid [illegible] coolants and [illegible] process units and steam generators. The [illegible] heating concept appears to [illegible].

The [illegible] Decomposer

The [illegible] on the SO_3 decomposition [illegible] Westinghouse (6), the Nuclear Research [illegible], Germany [illegible], General Atomic in San [illegible] Centre at Ispra, [illegible]. The theoretical effect of temperature and [illegible] on the SO_3, SO_2 equilibrium in the presence of water [illegible] available (10) and is shown in Fig. 3. At [illegible] atmospheres reactor exit total

Fig. 2. New concept for coupling the G.A. cycle to the TMR.

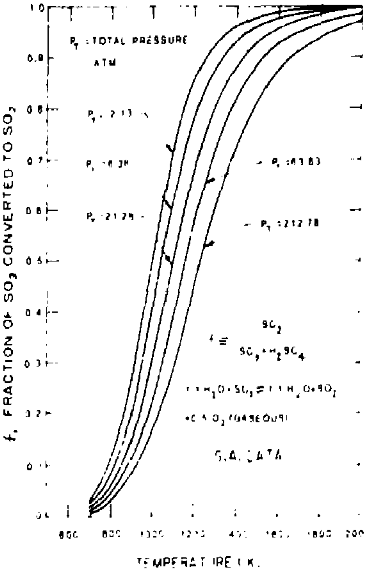

Fig. 3. SO_3-SO_2 equilibrium [illegible] (courtesy General Atomic Company).

pressure and 1250 K, the fraction [illegible] converted to SO_2 is around 84%. We have [illegible] for the SO_3 decomposer [illegible] eliminating the need for [illegible] problems in the decomposer [illegible] the TMR [illegible]. This compares to G.A.'s 74% [illegible].

Chemical Kinetics

Now that we have [illegible] the TMR to the [illegible], we need [illegible] the kinetics of the [illegible] design concept, we [illegible] to get SO_2 and [illegible] Joule-heated element [illegible] internal heat exchanger [illegible]. At 1250 K, as the estimated curve in Fig. 4 [illegible], alumina alone can provide [illegible] area, that no further precious metal or metal oxide catalyst need be used. [illegible] heating [illegible] was suggested by Oscar [illegible]. In our design, as we shall see later, we assume that [illegible] will be equally effective [illegible] the alumina shown in Fig. 4, and need not be catalytic.

There is also the problem of the back reaction of SO_2 and O_2 to SO_3 at 1250 K, as shown in a study by Rovner (10) in Fig. 5. Although his study was undertaken for a 2000 K SO_3 decomposition reaction, we believe (12) that his results can be used to estimate our 1250 K SO_3 decomposition case, as shown by the dashed curve in Fig. 5. We expect the conversion to decrease to around 77% owing to a back reaction resulting from a quenching rate of at least 300 K/sec in the cooling heat exchanger following the decomposer. We will discuss this quenching rate later in this paper.

H_2SO_4 Process Step Configuration

We have configured the H_2SO_4 process step in such a way as to keep the blanket at around 900 K and supply all of the thermal energy demand above 900 K from the electrical heating. As shown in Fig. 6 the multi-effect evaporator train is operated between 500

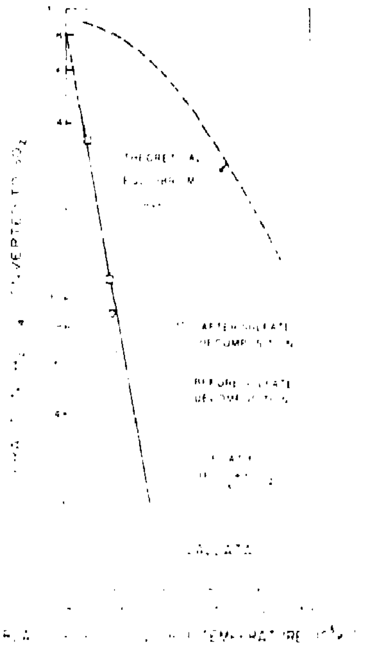

Fig. 4. Sulfuric acid decomposition on alumina substrate (Alcoa F 1-1 4 8.) (Courtesy General Atomic Company).

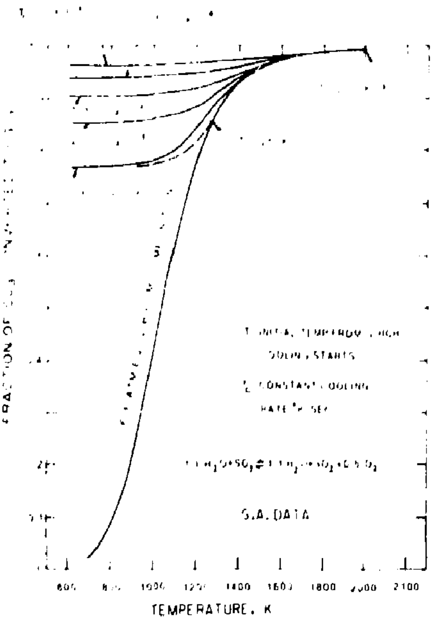

Fig. 5. Back conversion of SO_2 to SO_3 during cool down.

Fig. 6. Configuring the Sulfuric Acid Section for the Joule-Boosted SO_3 Decomposer.

and 680 K the H_2SO_4 is boiled at 680 K, the decomposer preheater is operated from 680 to 350 K and the SO_3 decomposer at 1250 K.

For this process configuration the thermodynamics for the H_2SO_4 SO_3 H_2O system can be used to generate an enthalpy-temperature curve as shown in Fig. 7 (1[illegible]). Note that in the temperature range of 700 to 1050 K no SO_3 decomposition is assumed to have taken place, whereas at 1050 K, as the process gas has passed from the preheater to the decomposer, the SO_3 decomposition in the decomposer is presumed to take place as the gas is heated from 1050 to 1250 K. The kinetic data in Fig. 4 help support this supposition since at a value of $10^3/T = 0.95$ (~ 1050 K), only about 3% of the SO_3 is converted to SO_2.

Our challenge now is to provide heat at the required temperature to the evaporators, boilers and decomposer preheater, and decomposer as shown in Fig. 6, using thermal and electrical energy from the direct converter and from the blanket and first wall. The fraction energy output available for process chemistry as well as the distribution between thermal and electrical energy from the direct converter is shown in Table 1 (2). We desire the lowest value of the TMR Q (ratio of fusion power out to injected power in) such that we can match the energy demands of the complete thermochemical cycle without raising the blanket temperature above 900 K. This value of Q of 15 gives a total fusion power output level of 3,000 MW_t from the TMR. After electricity is used for neutral beam injectors and ion cyclotron resonance heating, 2668 MW_t is available for the chemical process.

Table 2 summarizes the energy supply and demand. For this case, the overall cycle efficiency for producing H_2 is calculated as 48.9%. Parametric studies show significant improvements in cycle efficiencies with higher TMR Q and higher input temperatures to the decomposer.

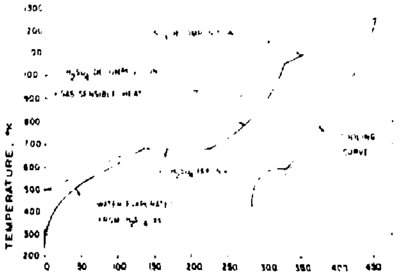

Fig. 7. Enthalpy heating curve for H_2SO_4/H_2O system.

Table 1. Direct Converter Energy Output Available for Process Chemistry (Injector and Direct Converter $\eta = 0.60$) Percentage of MW_{Fus}.

TMR Q	Blanket Thermal	D.C. Thermal	D.C. Elect. To Synfuel
5	95.09	16.00	-11.09
10	88.36	11.21	0.43
15	86.31	9.75	3.93
20	85.32	9.04	5.63
25	84.74	8.63	6.63
30	84.35	8.35	7.30
35	84.08	8.16	7.76
40	83.87	8.01	8.11
50	83.59	7.81	8.60

A longer term fusion goal is to ultimately operate on DD, in which case the particle flux into the direct converter *increases from 20% to about 50%, thus increasing* substantially the dc electrical power available to the process from 4% to around 30%. This would be sufficient to supply all of the electrical needs of the Joule-boosted decomposer instead of converting thermal energy to dc power with a loss in cycle efficiency.

Joule-Boosted SO_3 Decomposer

Last year we developed conceptual designs for the SO_3 decomposer involving a catalytic packed bed reactor (14-15), *a fluidized catalytic bed reactor (3), and* the catalytic cartridge heat pipe design (1). All of these design concepts made use of the higher temperature 1200 K Li-Na cauldron blanket design and made no special use of the unique properties of the direct converter.

The Joule-boosted decomposer is therefore unique, in that it uses electrical energy to heat SiC heating elements (suggested by Krikorian (3) to be commercially available and widely used today and believed by him to be compatible with the decomposing SO_3 vapors). Such *a decomposer design is depicted in Fig. 8. Large SiC* heating elements, 5.[illegible]4 cm in diameter and 3.04 m long are arranged vertically with the process gases fed in crossflow, very similar to commercial crossflow shell and tube heat exchangers. The ends of these heating elements are passed via insulators through the header sheets and are cooled by a coolant gas down to 600 K. These ends must be cooled in order to provide for the commercial, present-day aluminum impregnated electrical feed connection. This heat loss out the ends of the elements is negligibly small at around 1%.

In developing this design concept, we have sized the vessel and components. The SiC heating elements are proposed to be formed with small corrugations in the surface as shown in Fig. 9, to benefit the gas-solid heat transfer coefficient as we will discuss later. These elements are configured in a hexagonal array with a 6.1 cm spacing normal to the flow and [illegible] cm spacing in the flow direction as shown in Fig. 9.

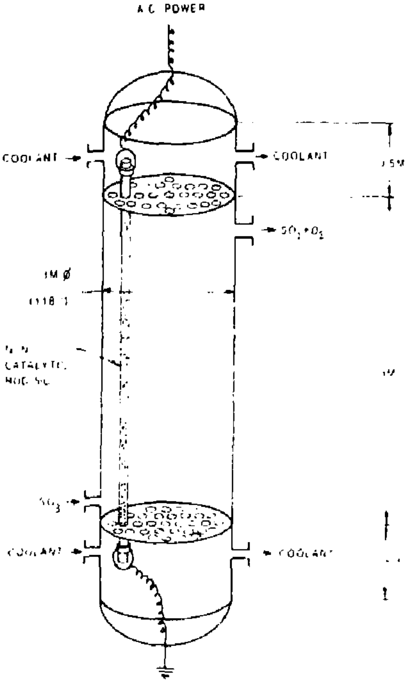

Fig. 8. Joule-Boosted SO_3 Decomposer.

These elements can be operated up to 130 kW m² (80 W/in²) at 1250 K. However, to achieve the longer life, beyond electrically heated furnaces and kilns, we have selected a lower 65 kW/m² (41 W/in²). We hope to achieve a 10 year life. At this flux, we need around 17,000 elements with a total extended surface area of 14,330 m² (0.84 m² per element). Using heat transfer correlations for staggered tubes in cross flow (14,15), we find that we can drive this decomposer at a gas to element surface ΔT = 73°K. Eight vessels are used. For this configuration the Reynolds number for *the process gases flowing cross flow to the SiC heating* elements, calculated at the minimum flow cross section is Re = 300,000, the Nusselt number is Nu = 410, and heat transfer coefficient is h = 120 cal/m²-s-K (92 Btu/h-ft²°F). At this rate of flow through

Table 2. Energy Supply and Demand Basis 3,000 MW_t TMR.

Unit	Temp (K)	Process Demand	Process Supplied	TMR Supply Direct Converter	TMR Supply Blanket
SO_3 Decomposer	950-1250	571 MW_t	0	94.5 MW_e	168 MW_e
Decomp. Preheater	680-950	720	0	0	0
H_2SO_4 Boiler	580	536	242	0	260
H_2SO_4 Evap.	380-680	773	664	0	109
Decomp. Recuperator	1250-800	0	720	0	0
Decomp. Cooler	800-418	0	1241	0	0
Remaining Units	418-680	702	0	0	702
Pumping Power	-	517	0	0	0

H_2 Product Chemical Energy Produced as Higher Heating Value = 285.2 kJ/gmol H_2.

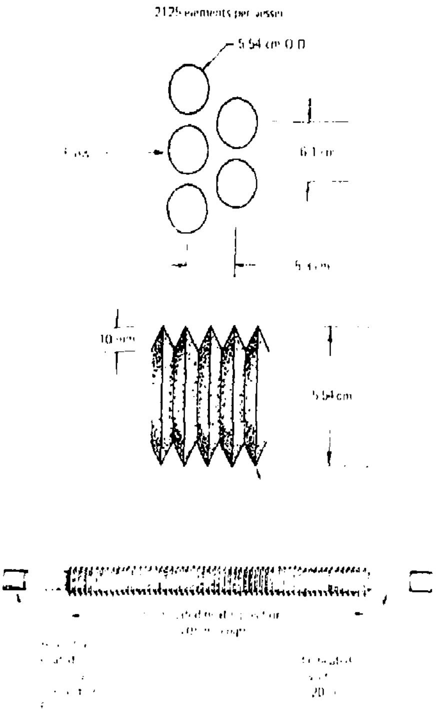

Fig. 9. Silicon carbide heating elements for the SO_3 *Decomposer*.

these eight vessels (i.e., 217 m^3/s), the pressure drop (in-out) would be 10.7 kPa (0.1 atm) and the pumping power is calculated at 23 MW_e total or 60 MW_t for blanket heat converted to electricity at 38% to run the blower.

We have selected this cross flow geometry for the decomposer heating elements because of significant heat transfer advantages. Cross flow around the 5.5 cm diameter elements is more effective (i.e., higher Reynolds and Nusselt number at a lower process velocity) owing to reformation and growth of the boundary layer and the separation and wake formation aft of the cylinder. These wakes provide turbulence which enhance *the heat transfer.* This heat transfer observation is based on an extensive experimental study (15) of staggered tubes in cross flow and we believe it is reliable.

One approach to estimating the SO_2/O_2 quench rate is to take the residence time of the process gas in the cooler. We assume the gas leaves the decomposer at 1170 K (i.e. 79 K below the 1250 K SiC element temperature) and no back reaction had time to occur as the SO_2 and O_2 is transported away from the SiC surface, through the thermal boundary layer into the gas. The *recuperator heat exchanger immediately following the* decomposer, we have designed as follows: six crossflow U-tube heat exchangers 2.6 m in diameter, 5 m in height. Operating at an LMTD = 139 K, with an overall heat transfer coefficient of h = 614 W/m^2-K (102 Btu/h-ft^2°F) and a tube surface area of 4500 m^2. There are about 480 U-tubes per unit which together handle the required 386 MW_t. For this design the quench rate would be 1400 K/s at a residence time of 0.36 s. According to Rovner's Fig. 5, this rapid quench rate would not cause a significant back reaction.

The eight SO_3 decomposer vessels lined inside with insulation can be fabricated out of Incoloy 800H to withstand the 0.7 MPa (7 atm) process pressure at a wall temperature of 1050 K and the corrosive $H_2SO_4/SO_3/H_2O$ gases. Using the criteria of Mori (16), adequate creep rupture strength (<1% creep in 20 years) at 12 MPa (1760 psi) can be provided using a 5 cm wall.

The cost of the SiC heating elements would be around \$17 million. The cost of these vessels can be estimated, using the Peters and Timmerhaus method (17) and based on a bare reactor vessel weight of 15 Mg (33,000 lb). The cost was estimated at \$100,000 each, fabricated out of Incoloy 800 (similar to 304 SS). *Installation of floating headers and the SiC heating* elements are expected to double the bare vessel cost to \$1.7 million, bringing the total to \$18.7 million. Using the "Lang method" (18) we arrive at a fixed capital investment of \$80 million including installation, instrumentation, piping, electrical, services, etc.; and engineering, contingencies, fees, etc. This is equivalent to around 21¢/GJ of delivered energy.

Comparisons to our other SO_3 decomposer designs on the same economic basis are shown in Table 3. We have used the "Lang method" (17) where the fixed capital investment is around 4.28 times the purchase price of the equipment.

Impact on Cycle Efficiency

Although we have not finished the complete heat integration of the H_2SO_4 section of the cycle with the other sections of the cycle, we can put together a rough power systems flow diagram as in Fig. 10. The major heat demand process units are found in the H_2SO_4 section, and the heat demands and electrical loads are consistent with our most current design concepts. The loads for the other sections are taken as identical to *those presented on the 1978 G.A. cycle flowsheet* (4), with the exception of the HI/I_2 distillation which is based on their very recent G.A. advances in process chemistry. There will be further shifts in the heat

Table 3. Cost Comparison with Our Other SO_3 *Decomposer Concepts.*[a]

Concept	Fabricated Cost M\$	Investments of Fixed Capital M\$	Contribution to Energy Cost ¢/GJ
Packed Bed	150	642	225
Fluidized Bed			
Sodium	32.8	140	49
Helium	52.3	224	78
Catalytic Cartridge			
Axial-Flow	46	197	69
Cross-Flow	26	111	39
Joule-Boosted	18.7	80	28

[a] Basis: 85% stream factor, 12% interest (I), zero equity, 20 year plant life (L), 3000 MW_t TMR, H_2 production at 4927 gmol H_2/s or 110 SM^3/s or 397,000 SM^3/h, energy production rate = 1.404 GJ/s at 47% cycle efficiency.

$$\text{Annualized cost, } A = \frac{I(1+I)^L}{(1+I)^L - 1} \times \text{Fixed Capital Investment}$$

$$\text{Contribution to energy cost} = \frac{L \times A}{\text{Energy Produced Over Life}}$$

Accuracy of Costs: ±50%

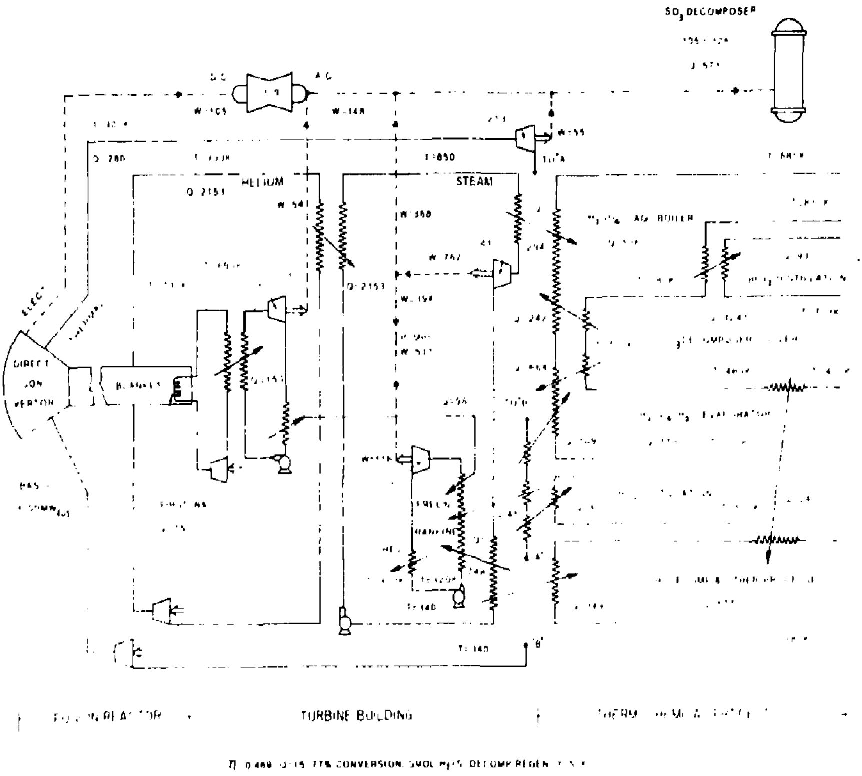

Fig. 10. Power systems configuration.

loads at several locations in the process, but they are expected to be minor. We have also assumed that around 320 K energy is rejected as low grade heat from the cycle. There are around ±5% errors in closing this first cut heat balance.

For the base case we have assumed a TMR Q of 15 and a decomposer input temperature of 950 K. We can make a rough prediction of the cycle efficiency from these heat and electrical power flows in Fig. 10. The total energy supplied to the process by the TMR is taken as the sum of the direct converter dc and the thermal output from the first wall, direct converter and blanket heat exchangers and is found to be 2668 MW. The H_2 production higher heating value is 285.2 kJ/gmol H_2 or 1305 MW_t at 4580 gmol H_2/s thus, the cycle efficiency is calculated at around 48.9%. This is the highest cycle efficiency obtained for this base case after a large number of efforts to configure the power systems with the minimum number of heat exchangers. It is apparent from this diagram that an effort was made to match up the temperature levels wherever possible and cascade the heat down to the lowest temperature levels in the process. Undoubtedly with further insight, small improvements can be made on this base case.

We have also done some parametric sensitivity case studies to examine opportunities for further reductions in cost. Some of the most significant cases are summarized in Fig. 11. Clearly the most important parameter is the TMR Q, since an increase from Q = 15 to Q = 25 increases cycle efficiency about 3%. Increasing the decomposer feed from 950 K to 1150 K increases the cycle efficiency about 2% We selected the lower temperature of 950 K as a "base case" since we feel the recuperating heat economizers on the decomposer can conservatively be made from Incoloy 800H, which may tolerate an 1100 K wall with corrosive process gases at 7 atm. Some innovative design techniques may allow us to raise the temperature.

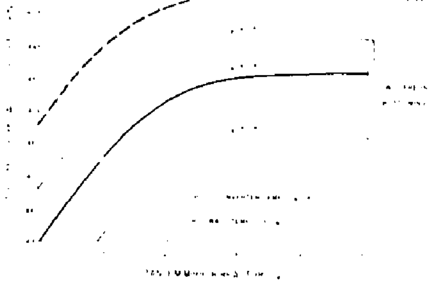

Fig. 11. Effect of TMR Q on cycle efficiency.

Also shown in Fig. 11 is the effect of removing the freon bottoming cycle and varying the blanket temperature. We have not as yet costed out the bottoming cycle, but suspect that the minimum cost plant may be achieved by operating the blanket helium at 1000 K and eliminating the bottoming cycle.

Also explored were cases for the cogeneration of H_2 and electricity for the grid. Cogeneration capitalizing on the unique features of the TMR direct converter appears really significant.

CONCLUSIONS

1. The Joule-heated SO_3 decomposer can be operated at 1250 K from available ac power from the direct converter and the blanket.
2. Silicon carbide heating elements are economically attractive and are believed to have compatible material properties for the H_2SO_4/H_2O system. They need not be catalytically active.
3. The Joule-boosted decomposer is a more compact and less expensive unit than any of the earlier concepts of packed bed, fluidized bed, or catalytic cartridge decomposer reactors.
4. The Joule-boosted decomposer offers the best possible safety isolation of the $SO_2/O_2/H_2O$ process gases from the liquid metal-containing blanket.
5. We have been able to develop a power-flow system layout that provides for utilization of various heat supplies and demands in order to obtain a cycle efficiency of around 49% with a low TMR Q = 15, 900 K helium from the blanket, a decomposer feed stream at 950 K, and a freon bottoming cycle.
6. Increasing Q to 25 raises cycle efficiency to around 51%.

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