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TITLE: PRESENT AND FUTURE STATUS OF THERMOCHEMICAL CYCLES APPLIED TO FUSION ENERGY SOURCES

AUTHOR(S): Lawrence A. Booth, S-10; Kenneth E. Cox, CMB-3; Robert A. Krakowski, CTR-12; and John P. Pendergrass, S-5

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PRESENT AND FUTURE STATUS OF THERMOCHEMICAL CYCLES
APPLIED TO FUSION ENERGY SOURCES

L. A. Booth, K. E. Cox,
R. A. Krakowski, and J. H. Pendergrass
Los Alamos Scientific Laboratory
Los Alamos, New Mexico 87545

Abstract

This paper reviews the status of current research on thermochemical hydrogen production cycles and identifies the needs for advanced cycles and materials research. The Los Alamos Scientific Laboratory (LASL) bismuth sulfate thermochemical cycle is characterized, and fusion reactor blanket concepts for both inertial and magnetic confinement schemes are presented as thermal energy sources for process heat applications.

Introduction

Since the advent of the industrial revolution in the late nineteenth century, the US industrial economy has relied on the use of hydrocarbon fuels and, as a result of this reliance, our end-use energy consumption has evolved to the point that 76% of our consumption is in energy forms supplied by oil and natural gas. Until the past decade we have had an abundance of these cheap, naturally occurring energy sources available; however, through the unabated use of these sources, we now find ourselves faced with an "energy crisis." This crisis is particularly acute because of the certainty that we cannot maintain our historical growth in demand for these fuels, and that eventually the supply of these fuels (specifically oil and natural gas) will be exhausted to the point that their use will be economically unattractive.

Thermonuclear fusion has the potential advantage of high power density and high temperature, and current program plans are directed toward demonstration of commercial feasibility near the end of this century. Therefore, fusion energy sources may become commercially available during the crucial period of projected energy source deficiency. The major problem with the use of fusion is that technology development is directed toward electric power generation, not toward the replacement of oil and natural gas as sources for the important areas of high temperature process heat and transportation fuels.

Therefore, it becomes essential to develop substitute hydrocarbon-based fuels for oil and natural gas to maintain our energy economy through this crucial transition period. Because of our extensive supply of coal, the most feasible means of providing this substitute is the production of synthetic fuels from this abundant resource. Production of synthetic fuels from coal is receiving great attention; however, the approaches poorly use the coal and require massive supplies of hydrogen.

The competition for producing hydrogen from fusion energy (aside from using other inexhaustible sources as primary energy) will be from the

standard coal gasification processes or from standard low-temperature electrolysis. The potential advantage over the standard gas gasification processes is the savings in coal of 50% so that as coal prices increase, the savings in operating costs can be made up by increased capital investment for the fusion/hydrogen production processes.

Even though the electrical efficiency of current low-temperature electrolysis processes are high (~70%), the overall efficiency is limited by the efficiency of electric power generation. Therefore, the efficiency of this method is limited to perhaps 30-35%. The most important factor in using electrolysis, however, is that the hydrogen production cost is primarily determined by electric power cost (and always must be higher on an energy basis). Therefore, any method of hydrogen production with overall efficiencies greater than by standard electrolysis will have important economic advantages.

The primary source of hydrogen today is the reforming of methane or naphtha, which is an example of an open-ended thermochemical cycle. Both heat and a chemical feed stock are required in addition to water. Electrolysis of water, which has been used in the past to produce hydrogen, can be considered a closed cycle in the sense that primary heat and water are the only feeds required.

The total energy value of the hydrogen used in the US is about 10% of the energy content of the methane that is used. Approximately 45% of the hydrogen is used for ammonia production, 4% for the refining of petroleum, and the remaining 10% for a variety of minor issues. For every gallon of liquid fuel produced from coal, at least one gallon of water is required for the hydrogen content. If the coal is to be gasified to high Btu gas, then two molecules of water are needed to supply the hydrogen for the methane. Clearly, production of hydrogen becomes an essential part of any synthetic fuel program.

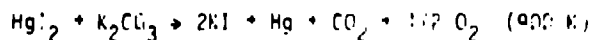
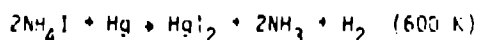
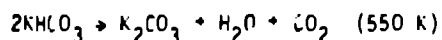
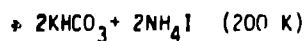
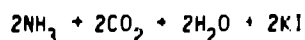
We have focused our attention on hydrogen production by closed thermochemical cycles because they require only water and a heat source. We have considered the status of current research on thermochemical cycles, which require temperatures in the range of 1100-1200 K; the need for research on advanced cycles at temperatures of 1500 K or higher; the need for high-temperature materials research; process characterization of the LASL bismuth sulfate cycle; and preliminary designs of fusion reactor blanket concepts.

Status of Current Research

There are a number of thermochemical water-splitting processes under development in the US and abroad. The funding for these processes is

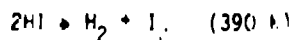
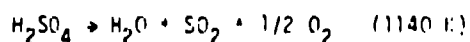
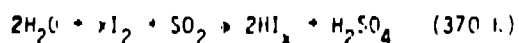
the US is presently provided by Department of Energy (DOE) and private organizations: three major processes are based on sulfuric acid decomposition and their thermal efficiencies are in the range of 40-50%. The development for these processes has progressed to the bench-scale level and engineering flow sheets have been designed for all three processes. There are other thermochemical processes under development in this country that are at a somewhat earlier stage of development. These processes involve the use of solids.

The ANL-4 process, developed at Argonne National Laboratory, is one of the few processes that has been identified as successful in splitting water. The process steps are as follows:



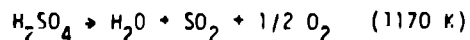
This cycle is not being actively pursued, primarily because of projected low efficiencies and the requirement for recirculation of a large variety of reagents including mercury, which is highly toxic.

The process presently under development at General Atomic Company (GA) requires only thermal energy and is described by the following reaction sequence.



In this pure thermochemical process, hydrogen-iodide and H_2SO_4 are produced in a low-temperature reaction from water, iodine, and SO_2 . The resulting sulfuric acid is decomposed in a high-temperature step to the SO_2 for recycle and the hydrogen-iodide is decomposed in a low-temperature step to yield hydrogen and regain iodine for recycle. The process is being demonstrated at the bench-scale level with a hydrogen production rate of 4 l/min. The presently quoted efficiency is 47%.

A process presently being developed by EURATOM in Ispra, Italy is a hybrid process requiring thermal and electrical energy. It is described as follows.



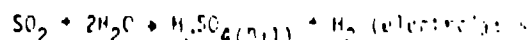
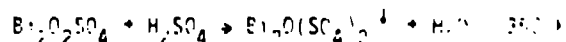
This hybrid process has been demonstrated at a production rate of 4 l of hydrogen per minute at the bench-scale level. A sulfuric acid decomposition loop at a scale of 1 ton H_2SO_4 /day is planned for FY 1980. Presently quoted process efficiency is 40%. The high-temperature step is the H_2SO_4 decomposition step, which requires a maximum temperature of 1140 K. The process also requires a significant amount of electrical energy for the electrolytic decomposition of HBr .

A process presently under development at Westinghouse is another hybrid cycle and is described as follows.



In this hybrid process, hydrogen is produced in a low-temperature electrochemical step where sulfuric acid and hydrogen are produced electrolytically and a high-temperature step where the sulfuric acid is decomposed to regain the SO_2 for recycle to the first reaction. The process is being demonstrated at the bench-scale level at a hydrogen production rate of 2 l/min. The presently quoted process efficiency is 41%.

The LASL bismuth sulfate process can be described as follows.



This hybrid process is in the laboratory development stage. The solid decomposition step has been demonstrated to be rapid, but the electrolysis step requires additional development. The presently quoted efficiency for the process is 50%. The potential advantages are: (1) achieving lower voltages for the electrochemical step; (2) minimizing energy required for drying solutions; and (3) minimizing corrosion problems in the dry system vs sulfuric acid cycles. Disadvantages are primarily those inherent in handling and transferring heat to solids. This example represents a family of potential solid sulfate processes using other metals.

The Need for Advanced Cycles and Materials Research

We believe that an inhibiting influence in the search for effective cycles has been the absence of a high-temperature heat source that could be matched to the chemistry. Because the GA, Westinghouse, and Ispra cycles require a

thermal decomposition of SO_3 to SO_2 and oxygen, efficient decomposition calls for a temperature of 1100-1200 K which is 200-300° higher than can be obtained from high-temperature gas-cooled reactors. The developing interest of the fusion program in synthetic fuels gives an impetus to the search for simpler, more effective cycles, and places the current SO_2 - SO_3 cycles on a sounder footing.

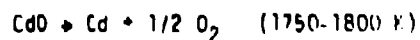
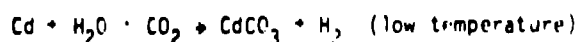
With current work in thermochemical cycles, the materials problems mainly are concerned with sulfuric acid evaporation and decomposition. Sulfuric acid is produced at ~50 wt% and needs to be concentrated to 98 wt% to avoid recycle of large quantities of water. Boiling H_2SO_4 solutions are highly corrosive and laboratory experiments have used quartz for containment. Current work involves SiC and other materials at temperatures of 500-800 K.

The problem with sulfuric acid decomposition is one of SO_2 contact with a high-temperature heat transfer surface. Here, a concerted materials research program is being conducted. Avoidance of the problem by direct contact heat exchange of the decomposing SO_3 with air in a ceramic tube may be a possible solution at temperatures in the range of 1000-1300 K.

Other problems at lower temperature involve containment of acidic solutions (HCl, HBr, and H_2SO_4) at moderate temperatures of 300-500 K. Here plastics may be of use. Solid sulfate or other solid systems may avoid certain of these problems but have the possible disadvantage of having to handle solids at high temperatures.

Although current engineering materials could not cope with 1500 K in the type of systems under discussion, it is within the realm of credibility to believe that materials and techniques can be developed to function reliably at 1500 K.

The principal reason for research on higher temperature (>1500 K) cycles is the potentially higher efficiencies attainable. An example of such a cycle has been proposed by LASS as follows.



This process has not been demonstrated in the laboratory, although the second two steps are known to proceed at reasonable rates. The first step may require catalysis to proceed at a reasonable rate. The ideal efficiency is 78%.

Process Characterization of the Bismuth Sulfate Cycle

A conceptual thermochemical process design has been developed for the bismuth sulfate cycle described above. For ease of analysis, the cycle was split into two portions, one including low-temperature operation and a high-temperature portion as shown in Fig. 1. A parametric analysis of the energy balance was done to determine the performance of the cycle and sensitivity of key process parameters over a wide range of operating conditions. For the reference parameters

indicated in Fig. 1, the overall efficiency, given by $\Delta H_{\text{H}_2} / \Sigma Q_{\text{TOT}}$, is 0.5.

Results from a sensitivity analysis to evaluate the effect of the three major system variables are shown in Fig. 2. These variables are the electrolyzer cell voltage, the endothermic heat requirement in the high-temperature portion, and the maximum stream temperature in the cycle. Held constant were the system pressure at 3 MPa, the mols of SO_3 removed at 1.0, and the mols of water entering the high-temperature portion at 5.0. In the case of the variation of cell voltage, the temperature was kept constant at 1475 K, and the endothermic heat at 285 kJ/mol H_2 . In the other two cases considered, the cell voltage was kept at 0.45 V (228 kJ/mol, heat equivalent).

The results show that the most important variables bearing on the cycle's efficiency are the electrolyzer voltage and the endothermic heat requirement. The effect of maximum stream temperature variation is important primarily because it varies the equilibrium yield in the $\text{SO}_3 = \text{SO}_2 + 1/2 \text{O}_2$ reaction and hence changes the composition of the gas mixture leaving the high-temperature portion of the cycle and the endothermic heat requirement.

Our earlier look at the variation in overall system pressure showed small changes in Q_{H_2} with pressure over the range 20-50 atm and thus not much variation from the point of efficiency. The major effect of varying system pressure in a gaseous system would be to change equipment sizes and structural requirements, thus influencing the capital costs of the cycle.

To obtain a cycle with high overall thermal efficiency, it is important to choose operating conditions that minimize thermal energy expenditure. In the case of the bismuth sulfate cycle, these conditions are fulfilled by the choice of a reasonably high maximum stream temperature (1475 K), high system pressure (3 MPa), low electrolyzer voltage (0.45 V), 1 mol of SO_3 removed per entering $\text{Bi}_2\text{O}_3 \cdot 2\text{SO}_3$, and a low number of mols of water entering the high-temperature portion of the cycle (<5 mols).

The high-temperature portion of the cycle is characterized by the flow sheet shown in Fig. 3. This portion of the cycle includes the bismuth sulfate drying step, the bismuth sulfate decomposition, and the sulfur trioxide decomposition steps. The fusion energy source supplies heat for endothermic decomposition of sulfur trioxide. When the $\text{SO}_3/\text{SO}_2/\text{O}_2$ mixture decreases to a lower temperature, the equilibrium shifts causing the back reaction, which gives off heat for the endothermic decomposition of $\text{Bi}_2\text{O}_3 \cdot 2\text{SO}_3$ in a "chemical heat pipe" mechanism. In this manner, primary heat from the fusion reactor is coupled to this portion of the cycle in a single heat exchanger. The other heat exchangers shown on the flow sheet are for internal heat recovery, that is, preheating incoming "cold" streams with exiting "hot" streams.

Entering $\text{Bi}_2\text{O}_3 \cdot 2\text{SO}_3$ is mechanically "dewatered" to 87 wt% solids and then is dried by direct and then indirect contact with a warmer stream of quenched $\text{SO}_2/\text{O}_2/\text{SO}_3$. Additional indirect heating is provided by an internal recycle stream of oxygen, by an $\text{SO}_3/\text{SO}_2/\text{O}_2$ stream, and by water vapor decomposition. Leaving the bottom of the drier, we have a vapor stream composed mainly of SO_2 .

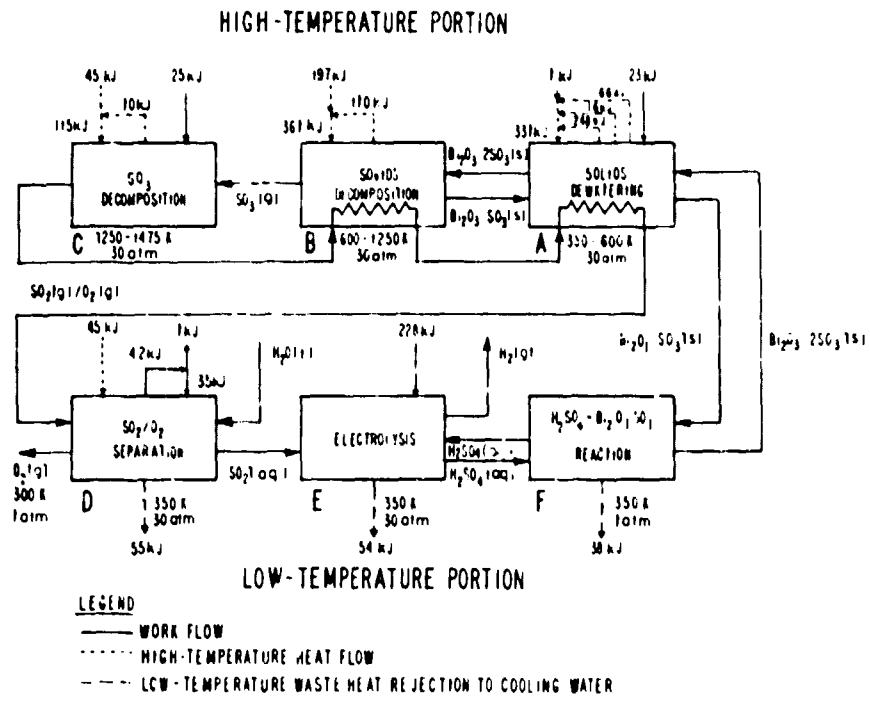


Fig. 1 Schematic of LASL bismuth sulfate hybrid thermochemical cycle.

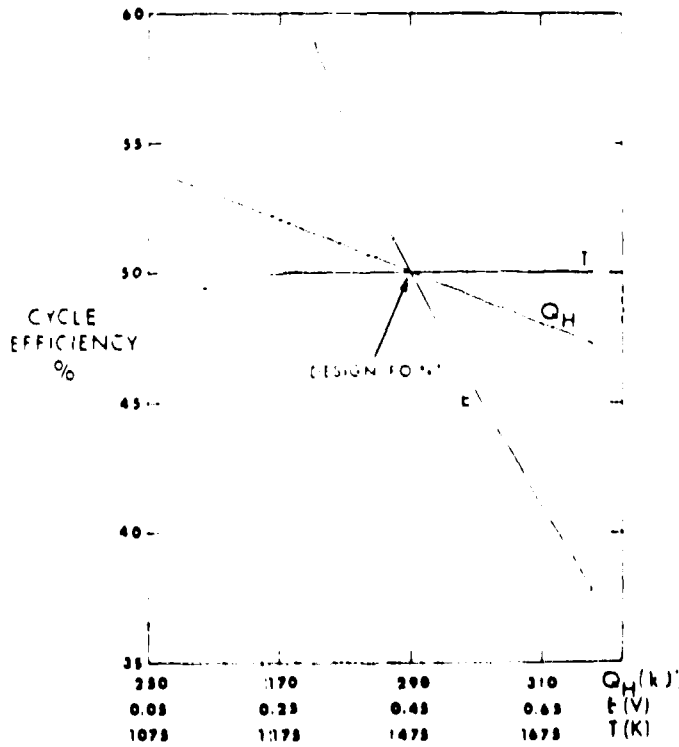


Fig. 2 Sensitivity of cycle efficiency to variations in electrolyzer cell voltage (E), endothermic heat requirement (Q_H), and maximum cycle temperature (T).

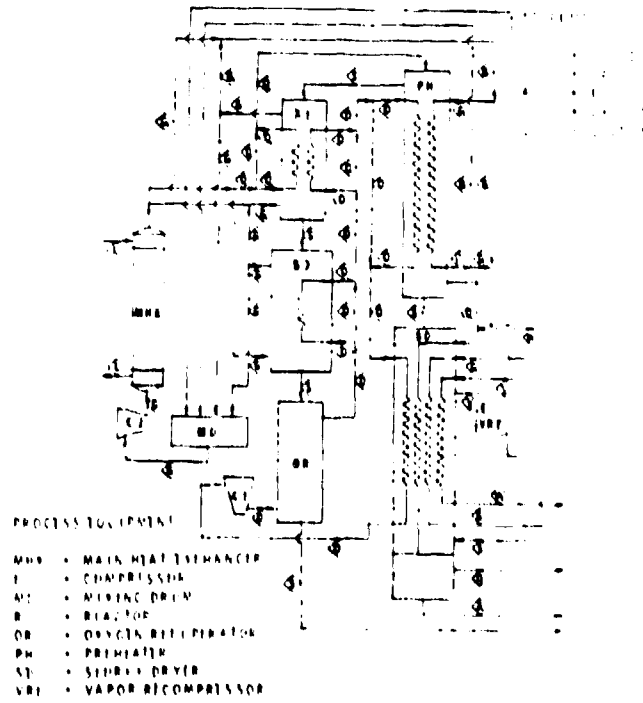


Fig. 3 Flow sheet of high-temperature portion of the bismuth sulfate cycle.

and O₂ and a liquid stream containing water and sulfuric acid. The dry solids exit the top of the drier vessel where they contact directly a quenched SO₂/O₂/SO₃ stream that comes directly from the SO₃ decomposer (primary heat exchanger) and is essentially unchanged in composition from its equilibrium at 1500 K and 3 MPa (30 atm). Heat exchange occurs between these streams raising the temperature of the dried Bi₂O₃·2SO₃ to ~800 K. Additional heat to raise the temperature of the solids is obtained from a recycle SO₂/O₂/SO₃ stream and from the recycle oxygen. The dried and heated Bi₂O₃·2SO₃ solids now are introduced into a series of decomposition reactors (two are shown in Fig. 3) where they come into direct contact with a SO₂/O₂/SO₃ gas mixture from the primary heat exchanger. We have assumed equilibrium conditions at the inlet and outlet temperatures of these decomposer vessels for the gas phase. In the decomposers, the solids are heated by two mechanisms—sensible heat exchange with the hotter gases and by the heat of reaction given up as some of the SO₂ and O₂ present shifts towards SO₃. This forms the heat recovery portion of the chemical heat pipe mechanism described earlier. This process dramatically reduces the gas flows from what they would have to be if only sensible heat were exchanged. As stated earlier, the decomposition reactors are solid-gas contacting devices. These units could have several configurations: fluidized beds, moving beds, or rotary kilns, etc.

In our design, we have considered the removal of 1 mol of SO₃ per mol of entering Bi₂O₃·2SO₃ as well as the removal of 1.33 mols of SO₂ to yield a product of Bi₂O₃·SO₃ or Bi₂O₃ (2/3) SO₃, respectively. The exiting solid material is cooled by the internally recycling oxygen stream before its discharge. This material is returned to the low-temperature portion of the cycle for subsequent contact with sulfuric acid and regeneration to the Bi₂O₃·2SO₃ starting material.

The low-temperature portion of the cycle is characterized in Fig. 4. Three major components of the process are included: the sulfur dioxide oxidation electrolyzers, the bismuth oxysulfate precipitation reactors and the units for separating Si₂ from O₂, and the extraction of energy from the oxygen stream by expansion to ambient conditions. In the electrolytic step, sulfuric acid is oxidized with the simultaneous formation of hydrogen at the cathode. In our design, we have assumed a slightly semipermeable, ion-conducting membrane that reduces sulfuric acid migration into the cathodic compartment of the electrolytic cell. The electrolytic cell design parameters have been chosen at: Cell Voltage: 0.45 V; Current Density: 2000 A/m²; Temperature: 350 K; Pressure: 3 MPa (30 atm); and Acid Concentration: 10-20 wt%. The effluent acid stream at 20 wt% is cooled by heat exchange before being reacted with the bismuth oxysulfate (Bi₂O₃·2SO₃) effluent from the decomposer vessels. These reactors produce a wet Bi₂O₃·2SO₃ precipitate that is returned to the high-temperature portion of the cycle. Some heat is liberated at low temperature in the reaction of Bi₂O₃·SO₃ with sulfuric acid. We estimate this amount of heat to be 38 kJ/mol, which is roughly half the heat release for the reaction of bismuth oxide with

sulfuric acid to form Bi₂O₃·SO₃. This heat serves to raise the temperature of the acid effluent from the bismuth oxysulfate reactors. The acid effluent then is introduced into a series of gas absorbers where it is used to separate SO₂ from the SO₂/O₂ stream leaving the decomposers. This operation recovers SO₂, which then is recycled with the acid back to the electrolyzers. The oxygen stream from the absorbers contains trace amounts of SO₂, which is further reduced by contact with the incoming water to the cycle. After final scrubbing, the oxygen stream is heated, expanded to recover useful work, and vented at close to ambient conditions to the atmosphere.

The principal choice of materials in the low-temperature portion of the cycle for components in contact with 350-400 K H₂SO₄ would be plastic-lined steel. At temperatures of 400-800 K, an acid brick may be used to line equipment. For the high-temperature portion of the cycle, we have to contain bismuth oxysulfates as well as dry SO₃, SO₂, and O₂. The high-temperature form of SiO₂ or a recently announced oxidation-resistant SiC sponge⁷ has capabilities to 1800 K. These problems, however, will require more detailed investigation.

High-Temperature Process Heat Fusion Reactor Blanket Concepts

A promising high-temperature process heat blanket system concept is illustrated schematically in Fig. 5.³ Fusion neutron energy deposited in boiling lithium is transported by vapor flow to and condensation on primary heat exchanger transfer surfaces. Lithium containment is achieved with thin refractory metal liners supported by actively cooled steel structural walls, with heat leakage from the boiling lithium to the steel structure limited by load-bearing insulation or by insulation pressurized with an inert gas to support the liner. Materials (TZ1, a gettered molybdenum alloy, or tungsten for the liner and carbon beam, ceramic foams, or multiple metal foils for the insulation) with the necessary properties have been identified.^{4,5} Detailed neutronics studies indicate that tritium breeding ratios of 1.5 combined with delivery of up to ~75% of total fusion reactor energy release as heat at 1500-2000 K are possible with this concept.

The lithium boiler concept has many attractive characteristics in addition to tritium self-sufficiency combined with delivery of a large majority of total reactor energy release as high-temperature thermal energy. Some of them are:

- thermal energy delivery as latent heat,
- low operating pressures,
- mechanical simplicity,
- efficient thermal energy transport,
- few geometrical constraints and compatibility with other reactor functions,
- self-pumping with gravity return of condensate,
- efficient dissipation by vapor bubbles of shock wave overpressures resulting from nonuniform neutron energy deposition characteristic of inertial confinement fusion by vapor bubbles,⁶
- temperature uniformity within the blanket that reduces material transport and eases design for thermal expansion,

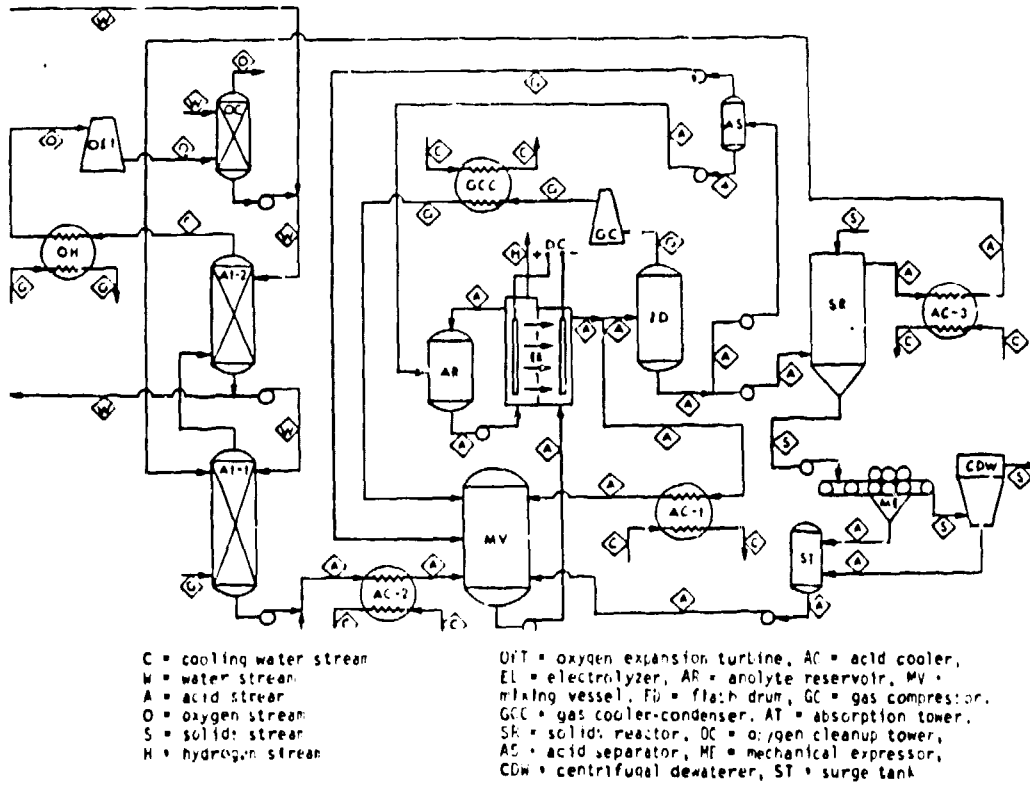


Fig. 4 Process flow sheet of low-temperature portion of the bismuth sulfate cycle.

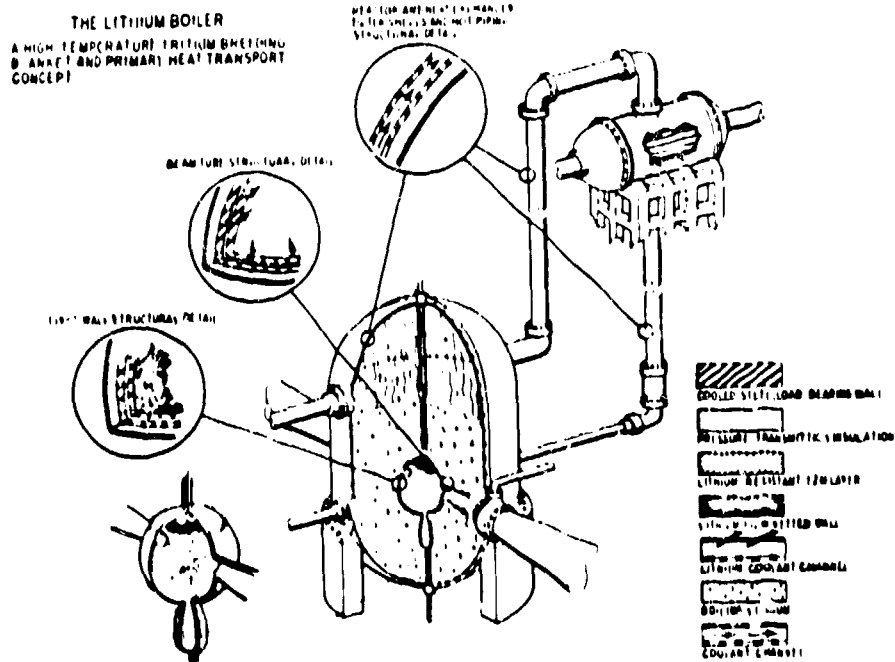


Fig. 5 The lithium boiler concept for inertial fusion application.

- requirement of only modest amounts of exotic materials of construction with no serious resource limitation, and
- no massive brittle components exposed to neutron irradiation.

Delivery of thermal energy as latent heat, that is, isothermally at maximum temperature, provides greater flexibility in satisfying process heat requirements. Lithium vapor pressures range from 1 atm at 1640 K to 5 atm at 2000 K, but can be adjusted up or down by adding, respectively, for example, other alkali metals or high atomic number neutron-multiplying metallic substances such as lead.

One potential drawback of the concept is that vapor volume fractions in the upper portions of MCF reactor lithium blankets may be so large that substantial and expensive increases in magnet bore may be necessary to provide room for condensed-phase lithium thicknesses adequate to intercept enough radiant energy that superconducting magnet shielding does not become a problem. However, our present understanding of boiling of liquid metals with volumetric energy deposition, as opposed to the more familiar boiling induced by heat transfer through surfaces, seems to be inadequate for accurate prediction of vapor volume fractions as a function of position in lithium boiler blankets. Our identification of this possible problem, which is not nearly so significant for ICF versions of the lithium boiler, is based on a relatively crude model of the boiling.

To circumvent this potential problem with MCF versions of the lithium boiler blanket concept, we have developed a nonboiling pressurized lithium blanket with many of the same design features.⁷ The principal differences between our pressurized concept, depicted schematically in Fig. 6 in a version adapted for incorporation into central solenoidal region modules of tandem mirror reactors (TMR), are:

- operation at pressures higher than the vapor pressure by an amount sufficiently great to ensure that boiling is suppressed everywhere in the loop,
- delivery of smaller amounts of total fusion reactor energy release at high temperature as sensible heat,
- requirement of mechanical pumping of liquid metals across magnetic field lines,
- internal structure required to distribute the liquid properly in the blanket, and
- lower volumetric flow rates but higher mass flow rates.

Operation at higher pressures means increased structural requirements for the blanket and primary coolant loop piping. However, freedom to select operating pressure levels above the boiling point means that pressure loading on primary heat exchanger transfer surfaces can be reduced by close matching of blanket operating pressure level to process operating pressure level. Our tandem mirror reactor version of this concept is calculated to deliver 61% of total fusion reactor energy release as sensible heat of liquid lithium at a maximum temperature ≥ 1500 K with a 200°C temperature drop around the primary loop and to achieve a tritium breeding ratio of

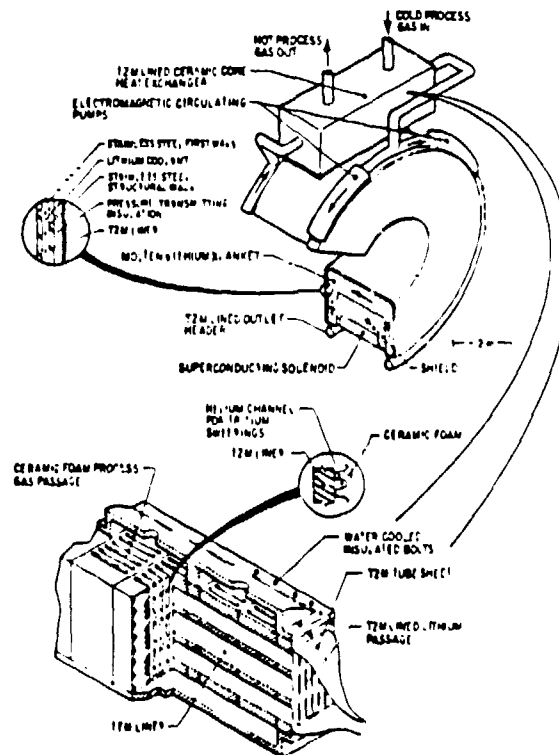


Fig. 6 The pressurized lithium blanket concept designed as a TMR process heat module.

1.46. Generous sizing of flow passages reduces required electromagnetic pumping power to only about 0.5 Mwe. The internal flow-directing structure is made of refractory metal alloy-clad ceramic.

In some respects, design of high-temperature primary heat exchangers represents a more formidable challenge than does blanket design because any loading on heat transfer surfaces must be born by materials at high temperature. As part of a study of fusion-driven thermochemical hydrogen production, we have developed a promising heat exchanger concept for heat transfer from lithium to oxidizing process gas streams at temperatures over 1500 K.^{7,8,9} A discussion of this concept, depicted in Fig. 7, will be used to illustrate some potential solutions to high-temperature heat exchanger design.

The crossflow plate-type design permits location of seals outside the high-temperature zone, ready assembly and disassembly for maintenance, and easier accommodation of differential thermal expansion. The lithium side is protected by thin refractory metal liners and ceramic structural material is an oxide for resistance to attack by the process gas. The oxide ceramic plus the helium-swept gap provides an effective barrier to tritium permeation. The helium gap can be pressurized to distribute loading on transfer surfaces if equalization of pressures on blanket and process sides is not possible. Double-walled construction can be used to provide additional protection against catastrophic failure and monitoring of the space between walls can provide early detection of leaks. Open cell ceramic foams in process gas passages reportedly also can provide significant additional structural support. Composite metal-

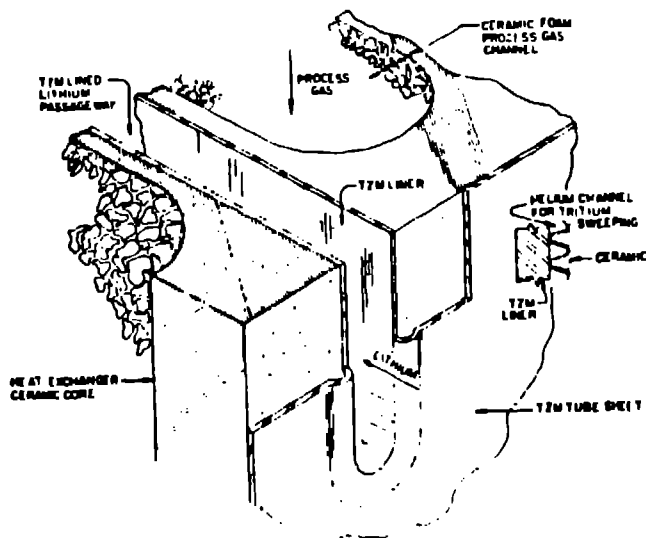


Fig. 7 Details of high-temperature plate-type process heat exchanger.

ceramic heat exchangers and ceramic foam heat exchangers are presently being developed.^{10,11}

Close coupling of primary heat exchangers with high-temperature fusion reactor blankets reduces the amount of relatively expensive piping required for heat transport, but must be weighed against reduced flexibility in coupling to processes. In addition, baffling to reduce neutron streaming into process fluids may be necessary with close-coupled exchangers.

Conclusions

Although the level of effort in thermochemical cycle research is low, several cycles based on sulfur trioxide decomposition show promise and will soon enter the pilot plant development stage. The use of solid sulfates can mitigate problems of handling highly-corrosive sulfuric acid at high temperatures, and the incorporation of electrolysis of dilute sulfuric acid (to produce hydrogen) increases the overall efficiency of the cycle. Although temperatures are limited to 1100-1200 K by current materials technology, we believe materials can be developed for oxidizing process streams at temperatures up to 1500 K. Availability of high temperatures (>1500 K) can lead to development of simpler and more efficient cycles.

Conceptual designs of fusion reactor blankets applicable to both magnetic and inertial confinement fusion have been developed to match the thermal requirements for any proposed cycle and to provide self-sufficiency in tritium breeding. These concepts appear feasible for circulation of lithium at temperatures >1500 K to a process heat exchanger.

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