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THULIUM HEAT SOURCE
IR&D Project 91-031

First Trimester Status Report
January 16, 1991

Carl E. Walter, Judith E. Kammeraad, John G. Newman,
Richard Van Konynenburg, and James H. VanSant

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Lawrence Livermore National Laboratory

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BACKGROUND

The goal of the Thulium Heat Source study is to determine the performance capability and evaluate the safety and environmental aspects of a thulium-170 heat source. Thulium-170 has several attractive features, including the fact that it decays to a stable, chemically innocuous isotope in a relatively short time. A longer-range goal is to attract government funding for the development, fabrication, and demonstration testing in an Autonomous Underwater Vehicle (AUV) of one or more thulium isotope power (TIP) prototype systems.

Our approach is to study parametrically the performance of thulium-170 heat source designs in the power range of 5-50 kW_{th}. (Originally we planned to study heat sources with up to 100-kW_{th}. Limiting our study to 50 kW_{th} maximum power seems appropriate in view of greater optimism about achieving higher thermal efficiency.) At least three heat source designs will be characterized in this power range to assess their performance, mass, and volume. We will determine shielding requirements, and consider the safety and environmental aspects of their use.

Our original conceptual design was a thulium oxide heat source cooled with heat pipes. The heat pipes would assure passive cooling of this "always on" heat source. Thin Tm_2O_3 wafers in a cylindrical stack would be interleaved with thick wafers of graphite. The stack(s) would be placed in a graphite block having suitable holes to accommodate them. This concept allows some of the beta radiation from Tm-170 to be stopped in low-Z material thus reducing bremsstrahlung radiation and consequently shield thickness. The concept is also compatible with irradiation of Tm-169 in the Fast Flux Test Facility (FFTF). The cross-section (resonance integral) of Tm-169 is so high that a thin wafer is required to prevent excessive flux depression during irradiation in a high, fast-spectrum neutron flux. Preliminary calculations indicated that about 50% of the Tm-169 placed in the reactor can be converted in a fast-spectrum reactor with a flux of 10^{14} n/cm^2-s in about two months. FFTF easily provides the appropriate flux characteristics.

SIGNIFICANT ACCOMPLISHMENTS

Other assignments have occupied much of the time of the Thulium Heat Source study team; nevertheless, we have made progress in our first trimester. We anticipate that we will be able to accomplish the goals of the study within the period and funding that was proposed. Because the study period is 9 months, it seems appropriate to provide this first trimester report of progress, a second report to coincide with mid-year reporting requirements (April), and a final report in July.

Significant accomplishments include:

1. We are considering modifications to our original concept that uses discrete disks of thulia (0.4 mm thick) interleaved with thicker disks of graphite (2 mm thick). The purpose of the graphite is to absorb the decay betas in a low Z material, provide good heat conduction, and (while being irradiated) a low neutron absorption material to space the thulia in the neutron flux. Preliminary calculations with the EGS code indicate that we can reduce the dose rate at the surface of the heat source by a significant factor if thulia and graphite are homogenously mixed. We will determine the best dilution ratio (currently the graphite-to-thulium atom density ratio is about 15). The choice of dilution ratio will strongly affect the conversion fraction of Tm-169 during irradiation, heat transfer parameters in the heat source, and the mass of the heat source. When the "best" dilution ratio is determined we will consider what material combinations should be used. This may lead to identification of the need to develop an isotope material form. Candidate materials for development could include hot-pressed $BeO-Tm_2O_3$ or $B_4C-Tm_2O_3$ (using B-11).

2. Westinghouse Hanford Co. has agreed to assist us by calculating the net Tm-170 production fractions for 30-, 60-, and 90-day irradiations in FFTF for target mixtures of $C-Tm_2O_3$ and $BeO-Tm_2O_3$. They will also determine how much Tm-170 could be produced annually in FFTF, assuming that FFTF were dedicated to long-term continuous production of Tm-170.

3. Our original concept uses heat pipes for heat removal from the heat source. We are finding that heat pipe performance in an adverse gravity field may be limiting. We found it expedient to write our own PC computer program to determine heat pipe performance limits. These scoping calculations used sodium as the working fluid. We are also considering forced convection as another means of heat removal. This may lead to a more compact design of the complete power system. We will continue to consider means of providing passive safety in a reversible manner with dedicated heat pipes that would function only during abnormal conditions (i. e. operation would continue after the abnormal condition is removed).

4. We have analyzed a promising, fail-safe, concept for the insulation surrounding the heat source. Multi-foil insulation in a vacuum annulus around the heat source provides adequate thermal radiation resistance to limit heat loss to less than 0.5% of the initial thermal power. If the normal heat source cooling system ceases to function, the heat source temperature will increase to a high but structurally safe value that is the melting temperature of a soft plug located in a line connecting a small helium reservoir, thus allowing the insulation annulus to be filled with helium. With helium present in the annulus, heat will be rejected at a reasonable temperature by radiation and convection. While continued operation of the power system would not be possible under this failed condition, the insulation could be refurbished after recovery of the system.

5. Research Chemicals, a Division of Rhône-Poulenc, Inc. has provided us with information on cost and availability of thulia (Tm-169). Their ore is xenotime (found in Malaysia) which contains about 1.3% Tm_2O_3. California ore (bastnasite) contains a much lower concentration of Tm_2O_3. We were advised that a requirement for 500 kg/y could be supplied at the end of 1992. This amount of material, if converted to 50% Tm-170, would provide a cumulative thermal power at the time of reactor discharge of 3400 kW. Since this appears adequate for wide use of thulium heat sources, we have not contacted other suppliers at this time. Current capacity of the Rhône-Poulenc plant is booked through 1991. Their plant has a theoretical capacity for producing Tm_2O_3 of 2000 kg/y. Cost could be as low as $3000/kg in quantities above 100 kg.

   The cost of producing the radioactive material will depend on the demand, the facilities available for production, and other factors. (In 1973, Pacific Northwest Laboratory estimated the cost of thulium-170 to be $136,000/kg. We are attempting to find out the basis for the estimate. Today the escalated cost would be $380,000/kg.)

6. To make our heat source study more relevant, we recognize that it is necessary to consider the performance of complete thulium isotope power (TIP) systems. To do this within budget, we have enlisted the aid of five well-known firms that are expert in specific areas of power conversion. We requested each firm to provide, at no cost to us, performance parameters of specific power conversion methods: (1) Stirling, (2) Brayton, (3) Rankine, (4) thermoelectric, and (5) HYTEC (a dynamic thermoelectric conversion concept). In subsequent proposals to the Navy or DARPA, we would invite these firms to participate in briefings, the proposal effort, and team with them in the development of prototype TIP systems. All five firms have indicated that they would provide us with assistance to a greater or lesser degree. Their responses are greatly appreciated. This will allow us to compare TIP systems using various power conversion methods and take into account their impact on heat source design. Each of the methods being considered has unique advantages and

disadvantages. Weighting of these attributes may be difficult until a specific application is identified; accordingly, we do not anticipate down-selection of power conversion methods during our study.

7. On October 24 we briefed Edward Mastal at DOE-NE-53 on our TIP activities. We had previously briefed James Turi, the former Director, Office of Special Applications, as well as Stephen Lanes, Deputy Assistant Secretary, and Earl Wahlquist, Associate Deputy Assistant Secretary, both in NE-50 and Robert Lange (NE-52) on our plan to study thulium isotope power systems for AUVs prior to our IR&D award. He suggested we contact Dr. Chryssostomidis in the Ocean Engineering Department at MIT as he had recently approached DOE for power systems for a particular application (see below).

8. We have talked on the phone with Dr. C. Chryssostomidis at MIT, as suggested by Ed Mastal of DOE. "Chris" is investigating an application for a Navy sponsor that requires small electric power systems for propulsion and data gathering in a number of AUVs. The cost of Pu-238 power systems is said to be $5-8M for 100 W_e. We provided him with some estimates of TIP system performance and cost. For example a 200-W_e TIP system might weigh less than 100 kg and occupy a reasonably small volume. The isotope cost should be less than $100,000. We will continue to follow up on this well-suited application for TIP systems.

9. We attended the DARPA briefing for prospective bidders for development of a fuel cell power system for the DARPA/Navy Unmanned Undersea Vehicle (UUV). The meeting was very informative. The power system performance together with the operational and vehicle constraints that are imposed, provide a realistic context to guide our study of TIP systems. The UUV is very large (1.1 m diam), apparently to accommodate the low-energy-density battery system currently available. The capability of the current power source, a Ag-Zn battery system, is 15 kW and 375 kWh with most of the power used for propulsion. The hull volume assigned to the power source is 2.59 m^3 and the battery components weigh 2.19 Mg. Note that this system provides 25 h operation at full power and that the mass and volume energy densities are 170 Wh/kg and 1.45×10^5 Wh/m^3, respectively. With fuel cells, the goal is to achieve 1000 kWh for the same power, mass, and volume, thus providing a three-fold improvement in endurance. The power requirement is a consequence of the large size of the UUV. DARPA/Navy's long range objective is to develop power systems that would fit in a 0.5-m diameter vehicle. The fuel cell developer is to provide a scalability study to accommodate a 5-kW power system in that size vehicle. A longer range objective of their fuel cell program is a ten-fold improvement in endurance over current battery capability. Our current rough estimate is that a 5-kW TIP system with 3000 h endurance (100-fold improvement) would weigh under 1 Mg and occupy a volume under 0.5 m^3 (with a diameter under 0.5 m), providing mass and volume energy densities better than 15,000 Wh/kg and 3×10^7 Wh/m^3, respectively.

10. In keeping with the expectation that research performed under the I R & D program be published, we have submitted an abstract of a paper for presentation (and inclusion in the proceedings) of the 26th Annual Intersociety Energy Conversion Engineering Conference (IECEC) to be held in Boston on August 4-9, 1991. We expect acceptance notification early in February.

- END -

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