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Infrastructure for Thulium-170 Isotope Power Systems for Autonomous Underwater Vehicle Fleets

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Abstract

The radioisotope thulium-170 is a safe and environmentally benign heat source for providing the high endurance and energy densities needed by advanced power systems for autonomous underwater vehicles (AUV). Thulium Isotope Power (TIP) systems have an endurance of ~3000 h, and gravimetric and volumetric energy densities of 3×10^4 Wh/kg and 3×10^8 Wh/m³, respectively. These energy densities are more than 200 times higher than those currently provided by Ag-Zn battery technology.

In order to capitalize on these performance levels with about one hundred AUVs in continuous use, it will be necessary to establish an infrastructure for isotope production and heat-source refurbishment. The infrastructure cost is not trivial, and studies are needed to determine its optimum configuration. The major component of the projected infrastructure is the nuclear reactor used to produce Tm-170 by neutron absorption in Tm-169. The reactor design should ideally be optimized for Tm-170 production. Using the byproduct "waste" heat beneficially would help defray the cost of isotope production. However, generating electric power with the reactor would compromise both the cost of electricity and the isotope production capacity. A coastal location for the reactor would be most convenient from end-use considerations, and the "waste" heat could be used to desalinate seawater in water-thirsty states.

Introduction

Proposals have been made in the past to utilize radioisotopes as energy sources for use in underwater power systems. A number of papers, published as early as the 1960s, considered using radioisotope decay energy for propulsion and other underwater mission power needs. One example is a 1968 paper: "A 7.3 kW_e Radioisotope Energized Undersea Stirling Engine" [1]. For various reasons, the few actual applications of radioisotope power have been limited to power levels of hundreds of watts. Recently, major technological and political changes have led us to believe that now is the time to develop a multi-kilowatt thulium isotope power (TIP) system suitable for wide use in autonomous underwater vehicles (AUVs). Systems containing thulium sesquioxide (Tm_2O_3), thulia, can meet stringent environmental and safety requirements, and will provide unequalled performance. The design and performance characteristics of such TIP systems have recently been presented [2].

Thulium occurs naturally as a single isotope, Tm-169, and is one of the heavier lanthanide rare earths. It is found in deposits of bastnasite, monazite, and xenotime ores in association with the other lanthanides, as well as yttrium and thorium. The half life of the Tm-170 isotope, which decays to a stable product (ytterbium-170), is 3086 h. Xenotime ore, particularly rich in thulium, is found in Malaysia. While both thulium metal and thulium have fairly high melting points (1820 K and 2650 K, respectively), thulium is refractory and offers greater chemical stability, with the possibility of operating at higher temperature for higher power conversion efficiency and better containment in accident scenarios. Thulium is one of the most stable oxides known. Natural thulium, at a price of \$3,000/kg, is available commercially in sufficient quantity and purity for TIP systems. Large quantities of Tm-170 can be obtained by irradiating natural Tm-169 in a nuclear reactor.

Thulium-170 Heat Source Design

We studied the thulium heat source in the configuration shown in Fig. 1. The heat source is cylindrical, and its length is approximately equal to its diameter. One end of the heat source is readily accessible for refueling. Various features at the other end provide heat-transfer, and integrate the heat source with a power conversion subsystem (PCS) through a heat exchanger. The heat-source material is in the form of thin (1.5-mm thick) hexagonal disks of thulium in several stacks, interleaved with similar disks of commercially produced graphite (for example, POCO AXF-5Q). The disks in each stack have a central hole to accommodate a heat pipe. Several stacks are nested together and placed in a thick-walled container, which provides both the proper containment for the radioactive material and an amount of shielding that yields an acceptable external dose rate. The inner surface of the container is suitably contoured to mate with the exterior of the heat-source stacks. Heat pipes protrude from one end of the heat-source container into an intermediate variable heat dissipator (VHD), which can be used to dissipate heat early in the mission (or at any other time, as needed) but prevents heat loss at the end of mission (EOM) and longer times. Table 1 lists the approximate characteristics for TIP heat sources at the beginning of mission (BOM) for three power levels: 5-, 25-, and 50-kW. Depending on the efficiency of power conversion, the electric power range of these heat sources (after one half-life) would be 0.25- to 12.5-kW. This electric power range would satisfy a number of undersea missions.

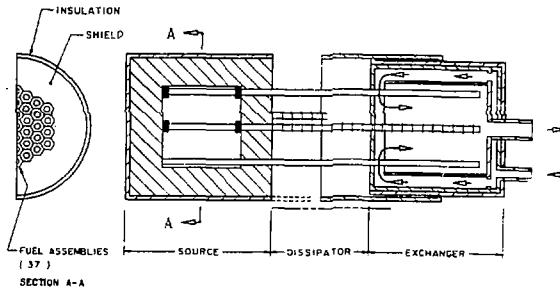


Fig. 1. Conceptual configuration of the TIP heat source integrated with a power-conversion subsystem.

Table 1. Heat-source dimensional and mass characteristics (unoptimized).

Parameter	Thermal Power (BOM), kW		
	5	25	50
Heat source			
Outside diameter, m	0.23	0.31	0.36
Outside length, m	0.25	0.36	0.36
Outside surface area, m ²	0.55	0.82	0.98
Number of heat pipes	7	19	37
Heat pipe length, m	0.23	0.54	0.80
Mass of Tm=170, kg	0.4	2.1	4.2
Mass (incl. heat pipes), kg	160.7	396.9	553.7
Volume, L	10.6	26.7	73.5
Variable Heat Dissipator			
Mass, kg	1.0	4.1	9.9
Volume, L	0.3	2.8	9.4
Heat Exchanger			
Mass, kg	5.0	20.7	49.4
Volume, L	0.3	3.0	10.7
Overall Length, m	0.31	0.62	0.89
Total Mass, kg	167	422	613
Total Volume, L	11	32	58
Energy density (@ 25% eff. EOM)*			
Gravimetric, Wh/kg	1.1E+04	2.2E+04	3.1E+04
Volumetric, Wh/m ³	1.7E+08	2.9E+08	3.3E+08

*For the heat source assembly only under these power conversion subsystem assumptions.

Power Conversion

Several power-conversion technologies can provide excellent performance [2] and further improvement is likely, as shown in Table 2. Besides overall efficiency, the other attributes that must be considered include reliability, maintenance, mass, volume, noise generation, and cost.

Table 2. Characteristic performance of suitable power conversion subsystems.

Power Conversion Technology	Thermal Cycle Peak Temperature, K (Growth)	Overall Efficiency, % (Growth)	Status
Brayton	1150 (1500)	30 (40)	Very mature
HYTEC	1300 (1400)	29	Developmental
Rankine*	925 —	21 —	Very mature
Stirling	1100 (1275)	42 (47)	Mature
Thermoelectric	825 (1350)	11 (24)	Mature

*Data shown are for toluene, not water.

AUV Missions

The conceptual TIP system can support undersea AUV missions requiring very high endurances. For example, three naval AUV scenarios are shown in Fig. 2 against a background of the inherent performance capability of a 50-kW_{th} (BOM) TIP heat source. The capabilities of three TIP systems are also overlaid in Fig. 2. The system's nameplate rating, such as 50-5C, describes the BOM thermal power in kW_{th}, the ECM electric power in kW_e, and whether the power is continuous, C, (as it is in this example), or whether variably higher (V) power exists before the EOM. Much longer mission times could be accommodated for missions 1 and 3 by using a TIP-50-5C or TIP-50-10C system, and a higher output power could also be provided for mission 2 with a TIP-50-10V system.

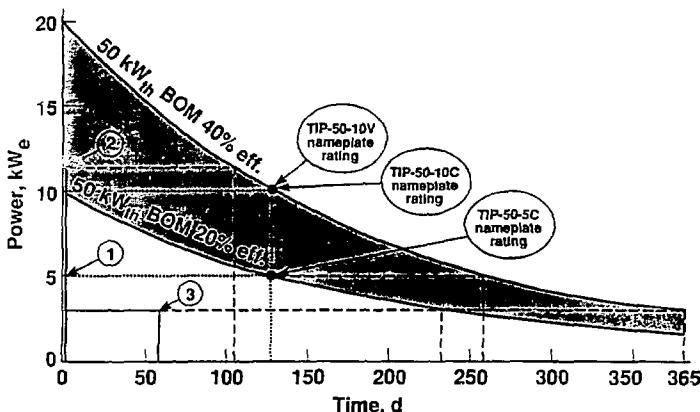


Fig. 2. TIP systems will have greater capability than their nameplate ratings indicate. Naval missions are easily accommodated: (1) search and survey, (2) tactical support, and (3) surveillance.

Production of Thulium-170

Thulium-170 is most readily produced in a thermal reactor operating continuously at a constant high flux. For efficient production of Tm-170, the target material thickness must be thin enough so that self-shielding effects are insignificant. In the late 1960s the Savannah River Laboratory studied Tm-170 as a potential heat source for power generation [3,4] by irradiating 1.5 mm-thick disks at two flux levels. At the higher level ($2.3 \times 10^{15} \text{n/cm}^2\text{s}$), a heat output of 2.5 kW/kg was obtained after 36 days of irradiation. No chemical or dimensional changes were observed, and at 2200 K the thulium disks were chemically compatible with a closed tantalum capsule. During the irradiation of thulium disks, a small amount of Tm-171 is also produced but by using 99.9% pure Tm-169, unwanted activation products will be kept within acceptable limits. Since the half-lives of Tm-170 and Tm-171 are 128.6 d and 1.92 y, respectively, after 50 days of irradiation in a thermal neutron flux of $1 \times 10^{15} \text{n/cm}^2\text{s}$, 26% of the

original Tm-169 would be Tm-170, and 4.9% would be Tm-171. The corresponding activity of Tm-171 would be ~4% that of the Tm-170, and the heat output (almost entirely from Tm-170) would be ~2.7 kW/kg of thulium.

The large-scale production of radioisotopes for various applications was envisioned in the early 1960s. It was recognized that a high flux would be required. A flux of $6 \times 10^{15} \text{ n/cm}^2\text{s}$ was demonstrated in a reactor at the Savannah River Laboratory [5]. The implications of radioisotope production, both in dedicated and dual-purpose reactors, were being assessed based on the assumption that 2 kg/y of productive neutrons would be available from a 1000-MW reactor operating at 90% of capacity. This production rate assumed 100% of the neutrons would be used to produce radioisotopes. Actually, fewer fission neutrons than this would be available for isotope production, since some will be absorbed in non-productive reactions, or will leak from the reactor. In our study, we assume that only half of the neutrons not required to sustain the fission process are absorbed in Tm-169 to produce Tm-170.

If 1 kg of neutrons were to be absorbed in Tm-169, they would produce 170 kg of Tm-170. Therefore, using the conservative factor of 0.5, a 1000-MW production reactor would produce 170 kg/y. This amount of Tm-170 would provide $6.2 \times 10^6 \text{ kWh}$ of decay heat during a single half life. Since some Tm-170 will be converted to Tm-171, and more will decay before being removed from the reactor, the Tm-170 yield is further reduced by a factor of ~0.8. We conclude that a special production reactor continuously operating at 3000 MW would have a conservative Tm-170 extraction rate of ~415 kg/y. We chose this power level as illustrative since it is representative of the size of current commercial power reactors.

With an extraction rate of ~415 kg/y, a sizeable fleet of AUVs could be maintained in operation at all times. Figure 3 indicates the number of AUVs at various power levels and power conversion efficiencies, that could be supported by one 3000-MW reactor. The curves in Fig. 3 are based on the assumption that each AUV in service would be refueled three times per year. The production capabilities at other reactor power levels to sustain a fleet of AUVs can also be inferred from the figure.

The compositions of the thulium targets withdrawn from the reactor depend in a strong manner on the effective neutron flux and the irradiation time. Estimates of these compositions were determined by solving the coupled Bateman equations describing the production and decay of isotopes with time in a neutron flux. The commonly accepted values of decay constants and absorption cross sections were used [6, 7], and the estimates for a thermal flux of $1 \times 10^{15} \text{ n/cm}^2\text{s}$ are shown in Fig. 4.

The variation of Tm-170 content with irradiation time for several irradiation flux levels is shown in Fig. 5. For different flux levels, there are profound differences in the peak conversion fraction of Tm-170, and in the times at which the peaks occur. A design objective for the production reactor should be to achieve as high a flux as is practicable.

A dedicated Tm-170 production reactor should be designed for easy Tm-169 target insertion and product removal. Ideally, this would be accomplished without having to shut down the reactor. It would also be desirable to be able to refuel the reactor while it is operating at full power. One way to accomplish these objectives is to use the mature production reactor designs that have been operated for over thirty years at the Savannah River Laboratory. However, new concepts should also be considered, which would make use of improved materials and provide even greater safety and a lower environmental impact. Also, new dual-purpose reactor

concepts should be considered as a means of reducing the cost of Tm-170.

Reactor concepts should be studied that allow full-power refueling, target insertion, and product removal. A concept that is proposed for consideration is a homogeneous reactor fueled by uranyl sulphate (UO_2SO_4) dissolved in D_2O as the moderator. The fuel/moderator mixture would circulate through a calandria to a low-temperature heat exchanger (under 400 K). By operating at a low temperature, difficulties with high-temperature corrosion can be avoided. (For example, there is a troublesome two-liquid phase region at the 550-K operating temperature needed to achieve reasonable efficiency as a power reactor.) Corrosion appears to be the only limiting feature of this design when operated at high temperature, and it leads to maintenance costs that result in a much higher (factor of three) cost of electricity than from conventional water reactor plants [8]. These economics led to discontinuing developmental work on the homogeneous reactor. However, corrosion should not be an issue in a low-temperature reactor. The tubes in the calandria would accommodate the target/product material, thus allowing insertion of the target and withdrawal of the product outside of the reactor primary coolant boundary. Adequate cooling of the target/product within the calandria tubes should be easily accomplished with a separate cooling loop. Reactor-fuel reprocessing could be accomplished on line, thus continuously reducing the fission-product inventory in the reactor to provide a high degree of safety.

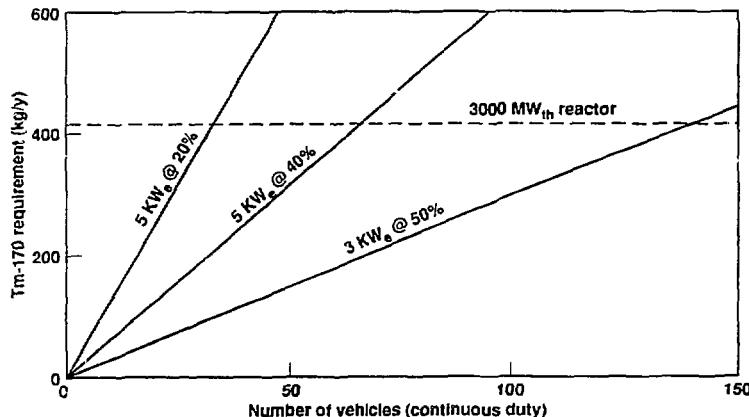


Fig. 3. A production reactor operating at 3000 MW_{th} would be able to support a large number of AUVs.

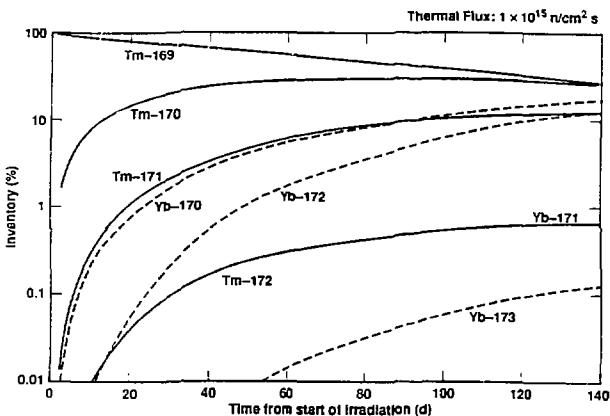


Fig. 4. The time-dependent composition of a thulium target during irradiation.

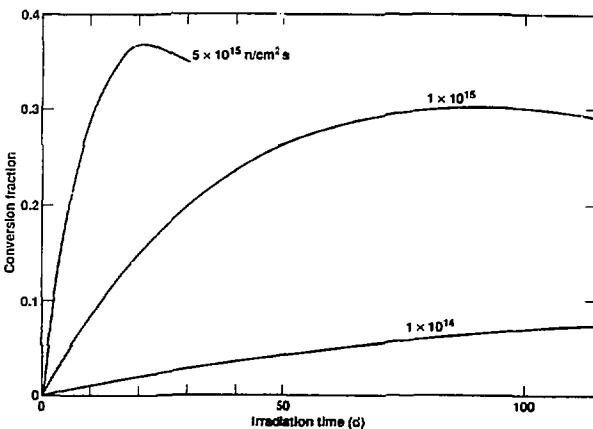


Fig. 5. The variation of the fraction of Tm-170 produced with irradiation times at three flux levels.

Waste-Heat Utilization

The low-temperature heat from the reactor could conveniently supply a desalination plant that would be operated in conjunction with Tm-170 isotope production. Several processes have been suggested for desalinating water, and a discriminator among them is the amount of energy required to separate the salt from the water. Processes such as reverse osmosis, using energy in the form of electricity, are among the most energy efficient. On the other hand, processes using thermal energy require a greater amount of energy. A measure of the energy required is given by the "performance ratio" commonly defined as: the number of pounds of fresh water produced per 1000 BTU of heat input.

In the dual-purpose plant needed to economically produce Tm-170, a great deal of "low temperature" waste heat is available, whereas electricity would need to be purchased. Accordingly, thermal processes such as multistage flash distillation (MSFD) or multiple-effect distillation (MED) are good desalination candidates for this application. Large desalination plants currently use the MSFD process almost exclusively. Typical performance ratios for MSFD and MED plants are 8 and 10 to 14, respectively [9].

In a recent study [9], MSFD and MED processes were evaluated and the latter was recommended for a dual-purpose (electric-power/desalination) nuclear plant. The recommendation was made on the basis of criteria requiring: (1) mature desalination technology, (2) successful commercial experience, and (3) best overall performance for an electric power/desalination plant. Although a recommendation for the desalination process to be used in an isotope production/desalination plant might be different, we proceed in this discussion with that recommendation.

Water with a total dissolved solids (TDS) content of less than 30 ppm would cost \$0.47/m³ [9] and could be sold competitively in water-thirsty California. This cost is leveled over a 30-year life. Blending the output with brackish water and improved desalination technology would further reduce this cost. Recently the Goleta Water District (near Santa Barbara, California) has considered importing water by ocean-tanker from Vancouver, BC at a cost of \$2.44/m³ [10].

In our case, the 3000-MW isotope/desalination plant would produce Tm-170 and fresh water at the annual rates of 415 kg/y and 4.7×10^6 m³/y, respectively. The leveled annual cost (including capital recovery) of operating this dual-purpose plant is estimated to be \$340 million, based on a recent cost analysis of a large nuclear-electric plant [11]. This cost would be offset by the sale of Tm-170 and fresh water. Various pricing strategies could result in more or less favorable prices for thulium or fresh water. If water were sold at the cost determined in reference [9], about two-thirds of the annual operating and capital costs would be offset by fresh water sales. The imputed cost of Th-170 would then be \$23/W_{th}.

Tm-170 Infrastructure

The continuous refueling of an AUV fleet will necessitate establishing a safe, environmentally benign, cost-effective, and smoothly running infrastructure. A block diagram for a potentially viable system is sketched in Fig. 6, which shows how material could flow in the TIP fuel cycle. All the activities within the heavy dashed box, except for reactor fission products storage, can be integrated at one facility. Activities that are outside the heavy dashed box do not involve radioactive material, with the exception of AUV operations. This arrangement would nearly

eliminate the transportation of radioactive TIP fuel material. If the reactor were located on the sea coast (as in California) the overland transportation of fueled AUVs could be avoided, and a substantial cost-benefit could result from the production of fresh water.

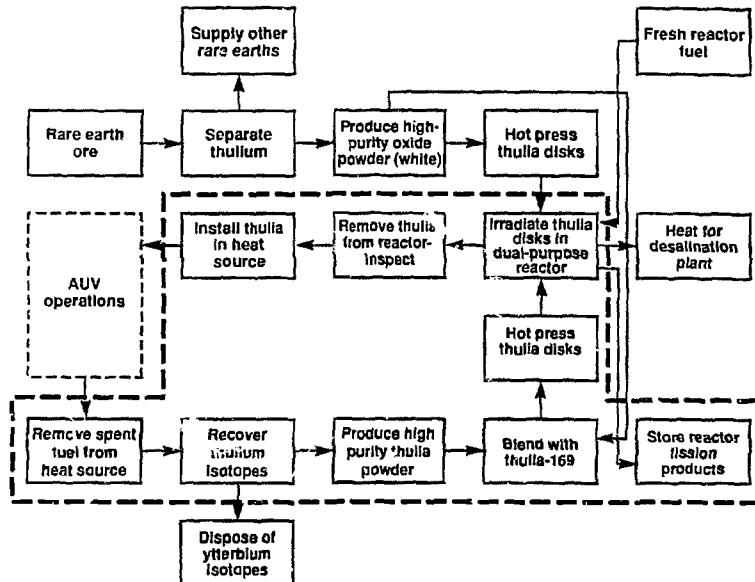


Fig. 6. Diagram of the infrastructure needed to fuel and refuel TIP systems.

If the production reactor(s) were owned and operated by private enterprise, their operation would be subject to Nuclear Regulatory Commission (NRC) regulation in a manner now practiced for power reactors. The NRC has issued a Master Material License to the Navy, under which the Navy issues permits to its appropriate operating units. The Navy is also responsible for inspections (self-assessments) and enforcing the applicable regulations. Furthermore, the NRC exercises audit and oversight functions to ensure that the conditions of the Master Material License are being met. The principal U.S. regulations that the Navy must meet are specified in the *Code of Federal Regulations*. International regulatory guidance is provided by the International Atomic Energy Agency [12]. The principal radionuclides present in a TIP heat source at EOM would be Tm-170 and Tm-171. Activated impurities do not appear to contribute significantly to the dose rate, and the spent source material (at EOM) would be classified as a Class B low-level waste [13].

Detailed design studies of the suggested infrastructure are needed to more accurately establish the costs of operating and maintaining a fleet of high-endurance "TIPed" AUVs. The benefits of such a fleet should greatly enhance Navy capabilities and accelerate undersea research and exploration.

Acknowledgements

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