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POWER APPLICATIONS OF RADIONUCLIDES

Justin L. Bloom
Martin Company
Nuclear Division
Baltimore 3, Maryland

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POWER APPLICATIONS OF RADIONUCLIDES

INTRODUCTION

For several years, the U. S. Atomic Energy Commission has sponsored an active program to provide relatively small nuclear power supplies for use in locations and under environments where more conventional sources of electrical power are unsatisfactory. Given the acronym SNAP (Systems for Nuclear Auxiliary Power), the program is divided into two major categories. The first is concerned with the exploitation of compact nuclear reactors, primarily for use in space vehicles. The second category covers the development of power supplies which derive their energy from the decay of radioactive isotopes and is the subject for discussion here.

To aid in the identification of the various SNAP programs, the odd-numbered designations (e.g., SNAP 3) refer to radioisotope systems, the even-numbered to reactor systems.

SNAP 1, SNAP 1A AND SNAP 3

In 1957, the AEC and the Air Force established a requirement for a radioisotope-fueled device which was to provide 250 watts of electrical power to the payload of an earth satellite. The first concept studied, termed SNAP 1, consisted of a radioisotope heat source which transferred its thermal power to boiling mercury; mercury vapor at high temperature then drove a small turbo-generator operating in a Rankine thermodynamic cycle. Not long after this program began, the avail-

ability of thermoelectric semiconductors for converting heat to electricity became a reality, and since there was some concern about the long term reliability of high speed rotating machinery, the program was revised to permit the use of thermoelectric conversion. The revised concept was called SNAP 1A, and was pursued into the hardware phase before the mission requirement was cancelled.

Concurrently, Martin began the development of a small proof-of-principal thermoelectric device called SNAP 3, in conjunction with Minnesota Mining and Manufacturing Company and Mound Laboratory. The SNAP 3 effort reached fruition in January 1959, when an operating unit was exhibited by President Eisenhower in the White House (Fig. 1). Decay of polonium-210 in the central core provided the thermal power which heated the hot junctions of the series-connected lead telluride thermocouples. About three watts of electrical power were produced with an overall thermal-to-electrical conversion efficiency of approximately 5%.

TRANSIT GENERATORS

SNAP 3 had been designed and fully tested to withstand the rigors of a space mission, although no requirement for its use existed at the time. With its successful operation demonstrated, the Navy, through its Transit satellite contractor, the Applied Physics Laboratory of Johns Hopkins University, decided to employ it to power part of the electronics system of the Transit 4 experimental navigational satellites.

Fig. 1. Artist's Concept of SNAP 3

Fig. 2. Transit Generator Being Tested in Laboratory

Fig. 3. Transit 4A Satellite and Generator

After some modifications in design and consequent additional testing had been performed, two fueled generators were delivered to the Navy.

Figure 2 shows one of these generators under test in the laboratory. Its principal difference from SNAP 3 is that it is fueled with plutonium-238, the relatively rare isotope of plutonium with a 90-year half-life. The use of Pu-238 ensures that a generator fueled with it will operate for many years with only a slight annual decrease in power due to decay.

On June 29, 1961, the first of these generators was launched into space as part of the Transit 4A satellite (Fig. 3)--the occasion of the world's first use of nuclear power in space. The generator has continued to operate successfully since that time and has provided more than 40,000 watt-hours of electrical energy to the satellite's radio transmitters. In December 1961, the second generator was launched aboard the Transit 4B vehicle, and it also delivered rated power for about six months until a failure in the external electrical circuitry cancelled its further effectiveness. Unfortunately, at about the same time, the entire power supply for the satellite failed when the parallel solar cell system was damaged by the manmade radiation belt resulting from high altitude nuclear weapons testing.

Upon this successful demonstration of the advantages of nuclear power in space, the Navy requested that the AEC provide full-scale nuclear power supplies for use in operational Transit satellites, and in a comparatively short period of time an entirely new 25-watt thermo-

electric generator, fueled with plutonium-238, was completely developed and tested. To date, three generators have been constructed and fueled, and now await launching by the Navy. One of the units is shown in Fig. 4. The series is designated SNAP 9A.

A feature of this type of thermoelectric generator should be noted at this point: plutonium-238 is ideally suited for space applications, since it is an alpha emitter. Little or no shielding is required to reduce the external radiation emission to levels where a fueled generator can be handled directly by personnel, or where the generator can be placed immediately adjacent to radiation-sensitive payloads.

Figure 5 illustrates how the Transit operational satellite will appear with its SNAP 9A nuclear power source.

SNAP 11

Another program that is being pursued into the hardware phase at the present time involves the development of a relatively short-lived generator which will deliver power to the NASA Surveyor spacecraft, a complex instrument package which will land on the moon. A model of Surveyor and its power supply are shown in Fig. 6. The generator, termed SNAP 11, is unique in that it will be fueled with the radioisotope curium-242. This alpha-emitting isotope has a half-life of only 162 days, but this is sufficiently long to provide design power throughout the 60-day operational life of the generator and spacecraft. Oak Ridge National Laboratory (ORNL) is now developing processes for the manufacture of Cm-242 and its subsequent encapsulation into heat sources.

Fig. 4. SNAP 9A Generator

Fig. 5. Transit Operational Satellite
with SNAP 9A

Fig. 6. Surveyor and Its Power Supply

The nuclear generator will provide power to the spacecraft during the long lunar night.

IMP

As a follow-on to the SNAP 9A program, Martin has begun the development of another generator, fueled with Pu-238, that will meet the requirements of the NASA Interplanetary Monitoring Probe, or IMP, vehicle. As shown in Fig. 7, two generators of 25 watts each are needed to provide the design power of 50 watts, because of restrictions placed on the generator configuration by the dimensions of a protective shroud during launch.

COMMUNICATIONS SATELLITE GENERATOR

While alpha-emitting isotopes are so desirable for space applications of nuclear power, they are also expensive and in short supply. We are therefore embarking on a program to develop a space power system which will be fueled with the plentiful fission product, strontium-90. Space generators fueled with strontium-90 are being considered only for missions where there would be an extremely high probability that re-entry of the space vehicle into the earth's atmosphere would occur only after the isotope had decayed to practically zero residual activity. With this restriction in mind, the most promising applications appear to lie in the field of communications satellites. The more sophisticated members of this family will orbit the earth at altitudes of the order of 5000 to 23,000 miles. Power levels in the range of 30 to 250 watts would be required.

Fig. 7. Artist's Concept of IMP

Fig. 8. Concept of Communications Satellite

Fig. 9. Generator Used in Axel Heiberg Weather Station

One of the significant problems to be faced in the utilization of strontium-90 for this purpose will be the minimization of radiation effects resulting from the bremsstrahlung produced as a consequence of beta decay. Undoubtedly, biological shielding will be required during the ground handling phase of readying the generator for launch. In space, shadow shielding and separation of the payload from the generator will be employed. In the artist's concept of a communications satellite shown in Fig. 8, the strontium-90 generator is mounted on a boom to increase the separation distance.

THERMIONIC CONVERSION

All of the systems described herein make use of the thermoelectric principle to convert heat to electricity. However, an alternative scheme, called thermionic conversion, is also being studied intensively. Although thermionic conversion is at present in the research and development stage, we can expect that it will become much more prominent in the future, since it has the potential of offering substantial increases in power-to-weight ratios and conversion efficiencies in comparison to thermoelectric conversion. As part of this effort, a thermionic diode to be fueled with curium-242 is under development. The demonstration device will be designated SNAP 13.

TERRESTRIAL SYSTEMS

In 1958, Martin Nuclear began a study of the use of strontium-90 as a heat source in thermoelectric generators for terrestrial applica-

tions under the sponsorship of the AEC Division of Isotopes Development. The first goal was to find a compound of strontium which would be stable at high temperatures, be fabricable by remotely operated processes, be essentially insoluble in fresh water and sea water, and contain as great a proportion of strontium as would be consistent with the other desired properties. The compound selected which most nearly met these criteria was strontium titanate, SrTiO_3 . It is a high density ceramic, melting at temperatures in excess of 3000° F, and exhibiting a rate of solution in sea water of the order of one microgram per square centimeter of exposed surface per day.

To demonstrate the beneficial use of strontium-90, the AEC then undertook the development of an integrated system which would utilize an operational prototype of a strontium-90 generator. An automatic weather station was held to be most representative of the concept.

NUCLEAR AUTOMATIC WEATHER STATION

With Oak Ridge National Laboratory providing the strontium titanate heat source, and with Martin Company designing and fabricating the weather station and power supply, the station was delivered to the U. S. Weather Bureau during the summer of 1961. On August 17 of that year, it became operational on Axel Heiberg Island in northern Canada, and it has broadcast regularly, without attendance, since that time.

The generator for the station is shown in Fig. 9. It weighs about 1600 pounds, of which more than 90% can be attributed to the lead

shielding required for bremsstrahlung attenuation. The 17,500 curies of strontium-90 which comprise the heat source are contained in about one pound of titanate and are encapsulated in the superalloy Hastelloy C. The generator container and its shield are both encapsulated in stainless steel for additional protection against attack by the environment.

The five-watt electrical power output of the generator is delivered to a nickel-cadmium storage battery through a dc-to-dc converter. The battery discharges intermittently to the load as the duty cycle demands.

Figure 10 is a close-up of the electronics package which receives weather information from the various sensors, stores it, and encodes it for transmission. It is completely transistorized, with the exception of the transmitter output tubes. Every three hours the station relays data on wind speed, wind direction, air temperature, and barometric pressure on two transmission frequencies. The data are received at Resolute, Canada, by a joint United States-Canadian team who feeds the decoded data into international weather networks.

The entire system, except for the antennas and weather sensors, is buried in the Arctic tundra within a steel tube 8 feet long and 2-1/2 feet in diameter (Fig. 11). This method of installation has several advantages:

- (1) The surrounding earth reduces the external radiation level to background.

Fig. 10. Electronics Package Used in Weather Station

Fig. 11. Cutaway of Weather Station Installation

Fig. 12. Ten-Watt Generator Configuration

- (2) Unauthorized tampering with the station is discouraged.
- (3) Thermal insulation provided by a styrofoam plug (in the top of the tube) and the surrounding earth enables the waste heat liberated by the generator to maintain the electronic components at temperatures of 50° to 70° F, regardless of the ambient air temperature.

SNAP 7 SERIES

A new class of generators, termed the SNAP 7 series, was initiated in 1960. They were to be more rugged, of greater power output, and capable of providing rated power for 10 years. Again, based on the use of strontium-90 titanate fuel, power levels of 10 and 60 watts were selected because of the applications planned for the generators. Two generators of each size were to be built.

In the 10-watt generator configuration (Fig. 12), four fuel capsules occupy the central region and are surrounded by 12 thermoelectric modules, each comprised of five thermocouples. The capsules and all external containment shells are fabricated from Hastelloy C. To reduce the weight of the generators, depleted uranium is employed in place of lead for shielding. A completed unit (Fig. 13) contains 40,000 curies of strontium-90 and weighs 1850 pounds.

The first generator which was completed was called SNAP 7C. It was shipped to McMurdo Sound in Antarctica in November 1961, and by February 1962 it was powering a Navy automatic weather station

Fig. 13. Completed Generator Loaded with Fuel

Fig. 14. SNAP 7A Generator in Coast Guard Buoy

Fig. 15. SNAP 7A Ready for Placement

at Minna Bluff. The station has operated intermittently since that time because of difficulties the Navy is experiencing with the station. The generator continues to produce its rated power.

The second generator, termed SNAP 7A, was delivered to the U. S. Coast Guard base at Curtis Bay, Maryland, where it was installed in a standard Coast Guard light buoy (Figs. 14 and 15). It was located at the lower end of the counterweight tube to facilitate removal of excess heat and to preclude damage in the event of collision by a ship. The generator powers a standard flashing light.

After about six months of operation at Curtis Bay, SNAP 7A began to show a loss in power output. Since the installation was of an experimental nature, the generator was easily removed from the buoy and returned to Baltimore for examination. It was found that the internal gas pressure of the generator had increased by about 0.5 atmosphere, causing the thermal conductivity of the insulation to increase, and resulting in a corresponding increase in unproductive heat losses. Analysis of the gas showed that it was principally hydrogen and helium. It probably results from desorption and decomposition processes in the fibrous inorganic insulation as the result of thermal and radiation effects. Reduction of the gas pressure to its original value restored the generator to its full electrical output. Metallic "getters" have been added to adsorb any further hydrogen formed. The buoy will be returned to duty in the near future.

The 60-watt generators of this series are called SNAP 7B and SNAP 7D. They are similar in concept and design to the smaller units described previously (Fig. 16) but their fuel inventory has been increased to 225,000 curies of strontium-90, contained in 14 capsules. Both generators have been completed. SNAP 7D is to be employed in the operation of a Navy boat-type weather station (Fig. 17). SNAP 7B will deliver power to a Coast Guard fixed light station at a position in the Chesapeake Bay.

Still another generator, SNAP 7E, was added to this program last year when the Navy requested that a power supply be furnished for deep-ocean use by the Navy Underwater Sound Laboratory at New London, Connecticut. Although its configuration is the same as that of SNAP 7A and SNAP 7C, only seven watts of power are required and consequently the fuel loading has been reduced to 31,000 curies. The other major change made is that the depleted uranium shield is replaced by a thick cast iron shield which also serves as a pressure vessel. In this application, SNAP 7E will energize an acoustic transducer (Fig. 18). A cross section of the power supply and other electrical components is illustrated in Fig. 19, and the completed hardware is shown in Fig. 20. The housing must be capable of withstanding immersion to a depth of 15,000 feet in sea water.

In this application, the generator charges a large electrolytic capacitor bank rather than nickel-cadmium batteries. The transducer is driven by discharging the bank periodically.

Fig. 16. Sixty-Watt Generators--SNAP 7B and 7D

Fig. 17. U.S. Navy Boat-Type Weather Station

Fig. 18. Arrangement of SNAP 7E and Equipment

Fig. 19. Cross-Section of Power Supply and Electronics Equipment

Fig. 20. SNAP 7E Completed

Fig. 21. SNAP 7E Being Lowered into Water

In September 1962, after a series of exhaustive hydrostatic pressure tests which demonstrated that the unit could withstand pressures of 10,000 psi, SNAP 7E and attendant equipment were transported by Navy vessel to a location off Bermuda, where an attempt was made to lower the system to its operating depth (Fig. 21). Much to the distress of all concerned, the transducer ceased operation when a depth of 12,500 feet was reached. The system was retrieved, and examination revealed that a leak had developed in the pressure vessel--probably due to stresses induced by the support cable which could not be simulated in land tests. The generator was unharmed, and work is in progress to make the necessary repairs and to undertake the installation again by the coming summer.

Since all of these systems are first of a kind and in many cases serve a technology that is new and unproven, it is not too surprising that some difficulties have been experienced.

MIXED FISSION PRODUCT GENERATOR

General Instrument Corporation has been developing a thermoelectric generator which is designed to make use of unrefined or so-called mixed fission products as the heat source. The fuel will be of very low power density in comparison to the refined fission products that have been employed so far. In all probability, it will consist of directly calcined wastes from the Purex process. A generator with an electrical heater simulating the isotope fuel has operated successfully

and a fueled generator is scheduled to be completed in the spring of 1964.

CESIUM-137 GENERATOR

Royal Research Corporation has been working on a generator fueled with Cs-137. The heat source has been fabricated by ORNL and consists of cesium borosilicate glass contained within a depleted uranium shield. When completed, the generator will be employed to power a deep-ocean seismic experiment conducted by Lamont Geological Observatory.

STRONTIUM-90 PROCESSING

Oak Ridge has prepared the fuel for all of the strontium generators described, with the exception of SNAP 7B. More recently, Martin has equipped a hot cell facility at Quehanna, Pennsylvania, for this purpose with AEC assistance, it being the first industrial facility to undertake large scale radiochemical operations of this nature (Figs. 22 and 23). In the five concrete-shielded hot cells (Fig. 24), about one million curies of strontium-90 per year can be processed into strontium titanate fuel pellets. Feed material for the process is supplied by the General Electric Company at the Hanford Works in the form of purified strontium carbonate, delivered in special casks (Figs. 25, 26 and 27). During the past nine months, Hanford has shipped 690,000 curies of strontium-90 to Quehanna in five shipments ranging from 94,000 to 170,000 curies each.

Fig. 22. Quehanna, Pennsylvania Facility

Fig. 23. Part of Hot Cell Operations Area

Fig. 24. Concrete-Shielded Hot Cells at Quehanna

Fig. 25. HAPO II Shipping Cask

Fig. 26. Cask and Protective Container

Fig. 27. Cask Assembly on Rail Car

Fig. 28. Schematic Process Flowsheet

In the Quehanna process, the solid strontium carbonate is removed from the shipping casks by dissolution in dilute nitric acid, an operation accomplished in Cell 1 (Fig. 28). Aliquots of about 10,000 curies are withdrawn from the stored nitrate solution and the strontium is precipitated as the carbonate in the presence of titanium dioxide in Cell 2. After washing by decantation, the precipitate is filtered in a ceramic filter crucible and is calcined in an electrically heated furnace to form the titanate. The particle size of the granular calcination product is reduced and the resulting powder is pressed into so-called "green" pellets, which are then sintered at 1400° C to produce the final fuel form.

From a purely chemical point of view, the process is quite simple. However, the entire operation must be conducted remotely, and no direct maintenance of equipment is possible without removal of the equipment from the hot cells and exhaustive decontamination. Another complexity that has arisen is that process development work carried out with inert materials proves to be inaccurate when adapted to radioactive materials. For example, if one wishes to calcine the carbonate-titania mixture at 1100° C, the furnace thermocouple may indicate that this temperature has been achieved, but the actual temperature of the radioactive powder will be considerably higher because of self-heating. Furthermore, the temperature of the powder will be a function of how it is distributed within its crucible. Since the production of high quality titanate pellets (ones with high material density, accurate dimen-

sions and free of cracks or fissures) is extremely sensitive to process variables, it has been necessary to repeat the earlier process development work with fully radioactive material.

All chemical processing of the isotope is done within a stainless steel containment box within the hot cell enclosure. Use of a double barrier compounds problems of egress and entry but affords a substantial degree of protection against the release of radioactivity.

Since the facility is located on private property, all radioactive wastes which are generated must be removed in shielded containers and transported to Oak Ridge for burial. To reduce the costs of waste disposal, the volume of liquids which enter the process, and which ultimately become radioactive waste, must be restricted. To date, only 40 or 50 curies of strontium-90 in the form of liquid waste have been lost, but losses in the form of dry waste have been substantially greater. Dry waste is generated principally as the result of radiation damage to plastic boots which cover the master-slave manipulators. The boots must be replaced frequently, and the damaged boots are then packaged for burial at Oak Ridge.

Fuel for the SNAP 7B generator was made at Quehanna and the generator was loaded there on January 15, 1963. Since that time, another 200 kilocuries of titanate have been processed into pellets for future generator loadings.