Production of Pu-238 Oxide Fuel for Space Exploration

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Abstract

The Savannah River Site (SRS) made significant contributions to NASA space missions via the heat source programs that provide electrical power to satellites traveling outside of earth orbit. Electrical power was supplied by converting heat from the radioactive decay of plutonium-238. The Site produced the Pu-238 in its reactors, separated the plutonium in the canyon facilities, and, for the Multi-Hundred Watt and General Purpose Heat Source programs, formed plutonium oxide into pellets and encapsulated the pellets in the Building 235-F Plutonium Fuel Form Facility (PuFF). Technological innovations were behind the success of each of these steps in production. Using Pu-238 produced at SRS has allowed U.S. satellites to explore the solar system and beyond, sending remarkable pictures back to earth. Plutonium produced at SRS has powered 26 space missions. This report particularly addresses the technology for fabrication of plutonium oxide pellets and encapsulation of these pellets. Plutonium oxide pellets produced and encapsulated at SRS are now powering the Galileo satellite presently circling Jupiter and the Ulysses satellite presently in a polar orbit around the sun.

Introduction

Early in the U.S. space program, scientists recognized that an efficient source of power was needed for satellites. Batteries had the disadvantages of being very heavy and had lifetimes that were too short for deep space missions. Solar cells were in their infancy and could not operate at great distances from the sun. Technology to convert thermal energy into electricity was available, and the decay heat associated with radioactive materials could be the source of energy. Coupling radioactive decay heat with a thermoelectric converter became the power source of choice for satellites, particularly those that need to operate where solar energy is not plentiful.

Plutonium-238, produced and packaged at SRS, was selected as the heat source for thermoelectric power for satellites used for deep space missions. During the late 1950s, the Atomic Energy Commission requested that the Savannah River Site produce Pu-238 as a heat source for this application. To accomplish this new task, a massive interdisciplinary effort was required. Neptunium-237, the precursor isotope to produce Pu-238, was recovered from existing SRS processes where it had been produced as a byproduct in the reactor irradiation of uranium. The neptunium-237 was fabricated into targets and irradiated in SRS reactors to form Pu-238. The Pu-238 was separated from neptunium and fission products and processed to plutonium oxide (see “Development and Performance of Processes and Equipment to Recover Neptunium-244 and Californium-252” in this proceeding).

Plutonium-238 is currently supplying power for the Galileo spacecraft orbiting Jupiter and exploring this planet and its four moons; the Cassini spacecraft, which is on its way to a similar mission near Saturn; and the Ulysses spacecraft, which is in polar orbit around the sun. Long-term, deep-space missions, such as

Galileo, Ulysses, and Cassini, require a nuclear source to provide the electrical power needed to operate the instruments on board the spacecraft. Similarly, smaller radioisotopic heater units provide localized heat for electronic packages on the spacecraft.

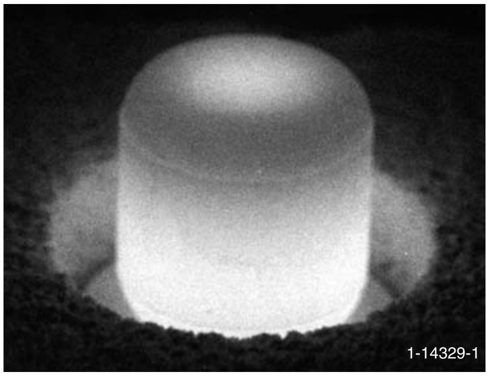
The Unites States has used, and continues to successfully use, radioisotopic thermoelectric generators (RTGs) to supply electrical power for deep-space missions. The RTGs consist of a nuclear heat source and a converter to transform the heat energy from radioactive decay into electrical power. Plutonium-238 serves well as the heat source because of its high power density (0.5 thermal watts per gram from alpha radiation), and its half-life of 87.4 years provides reliable and relatively uniform power over the lifetime of most NASA missions. The RTG is also a very reliable power source because it has no moving parts. All onboard electrical power to operate the cameras, collect the data, and relay information to Earth originates from the Pu-238 fuel produced at Savannah River.

Significance for SRS

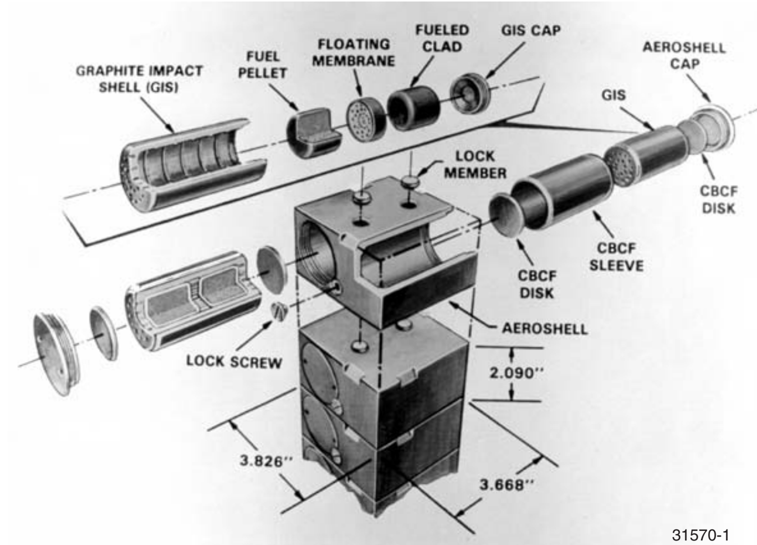
Major scientific endeavors in space receive great benefit from technology at SRS. This technology produces and recovers Pu-238, processing the isotope into oxide, forming the oxide into pellets, and encapsulating pellets for long-term, high-temperature applications in U.S. space programs. Plutonium-238 produced and recovered at SRS supplied electrical power for all of the NASA deep-space probes, as well as the highly successful Apollo manned missions to the moon, the Galileo mission to Jupiter, the Ulysses mission around the sun, and most recently the Cassini mission to Saturn. In addition, data from instruments left on the Moon by the Apollo astronauts was transmitted to Earth for many years using electrical energy generated from the heat sources. The attention-getting Voyager 2 mission, which resulted in major discoveries about Jupiter, Saturn, Uranus, Neptune and their respective moons, was able to transmit, as a direct result of technology developed at SRS, information even after a 22-year, 15-billion-mile space odyssey.

The Galileo and Ulysses missions are particularly significant to SRS because these spacecraft use Pu-238 that was processed into pellets and encapsulated at SRS. The General Purpose Heat Source, containing 72 SRS encapsulated pellets, supplied the electrical power for these missions, which are ongoing at this time. The Galileo satellite (see Figure 1) has completed its initial objectives of exploring Jupiter and its four major moons. Additionally, it took the first photographs of the Earth and Moon together ever taken by an unmanned spacecraft, provided the first close-up photographs of an asteroid, and photographed the collision of comet Shoemaker-Levy 9 with Jupiter. The Galileo mission was extended for further studies of Jupiter's moon Europa. The Ulysses mission to fly over the poles of the sun was completed and provided a greater understanding of the behavior of sunspots, solar flares, solar x-rays, and solar radio noise. The Ulysses mission was also extended to investigate the high latitude properties of the solar wind. These extensions demonstrate the reliability of the Savannah River-produced Pu-238 pellets to provide power for extended space missions.

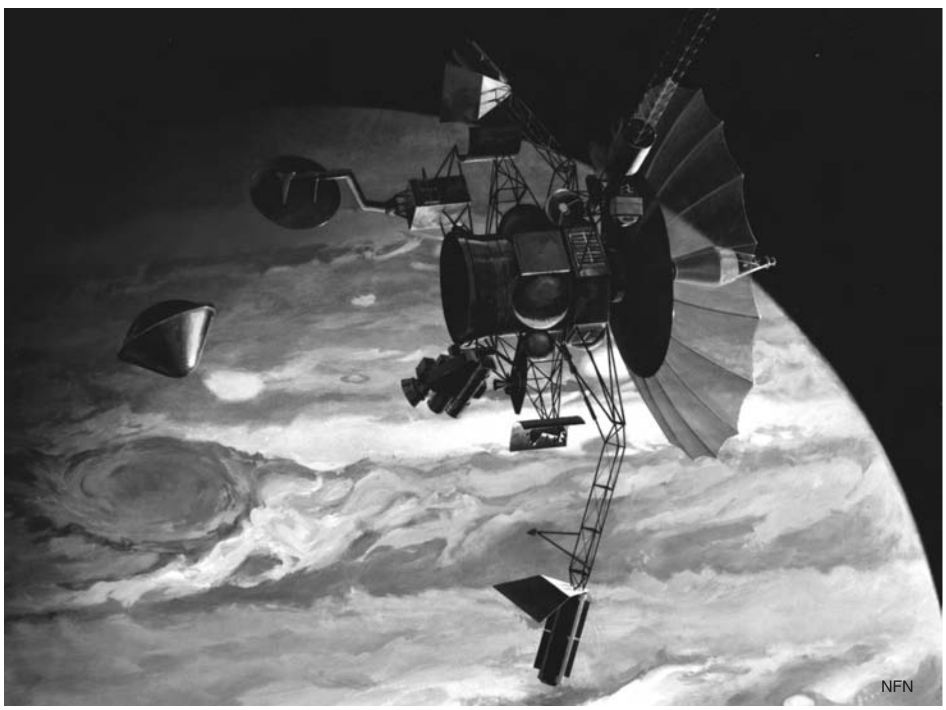
The Savannah River Site has played a major role in the U.S. space program. The successful use of Pu-238 in space allowed ventures outside of earth orbit that could not have been done without this reliable, long-lasting power source. The successful space missions that have occurred in the past and continue today were possible because of developments at SRS, beginning early in the history of the Site. Technologies used to develop Pu-238 for space missions include reactor physics, chemical separations, ceramic engineering, joining technology, materials engineering, and mechanical and electrical design. The extensive knowledge of the planets, the sun, and the moon that are available today are, in part, possible because



A. Photograph of General Purpose Heat Source $^{238}PuO_2$ pellet glowing from its own heat



B. Assembly of SRS $^{238}PuO_2$ pellets into General Purpose Heat Source module



C. Galileo satellite orbiting Jupiter (artist conception). GPHS Radioisotopic Thermoelectric Generators (on lower and upper booms) each contain 72 encapsulated $^{238}PuO_2$ pellets produced at the Savannah River Site using pellet fabrication and encapsulation processes developed at Savannah River.

Figure 1. Galileo mission to Jupiter powered by the General Purpose Heat Source, 1 of 26 space missions using SRS plutonium to provide power

of the ingenuity and devotion to scientific technology shown by many at SRS over the past 50 years.

Processing Pu-238 into Pellets

The preferred chemical form for a plutonium heat source is $^{238}PuO_2$, a ceramic (Rankin 1982). This face-centered cubic oxide offers excellent chemical stability, a high melting point (>2450°C), and chemical compatibility with its container material, an iridium alloy. The absence of phase changes in this material facilitates fabrication and enhances the integrity of the heat source during processing and use at an operating temperature of about 1350°C. A fabrication process, developed at the Savannah River Site, produced 150 g cylindrical pellets, approximately 2.7 cm by 2.7 cm, each of which generated 62.5 watts (thermal) at the time of production. Seventy-two of these pellets are contained in a single RTG that produces 285 watts of electrical power. The RTGs are compact (0.42 m in diameter by 1.13 m in length) and low in weight (55.5 kg), two requirements for efficient space systems. Two RTGs supply all the electrical power in the Galileo spacecraft.

The fabrication process for the cylindrical pellets consists of hot pressing a blended mixture of sintered $^{238}PuO_2$ granules prepared from calcined plutonium oxalate powder. Hot pressing provided the dimensional control needed and produced nominal pellet density of 84.5% theoretical. Both the granule sintering and hot processing conditions required close control to minimize cracking and to eject the pellet from the hot press die. A uniform pellet density and the distribution of large intergranuar porosity in the microstructure provide for the dimensional stability and the release of the decay helium (from alpha particles) required at the elevated use temperature.

A process suitable for pellet production was developed at SRS. This development was initially based on experiments at the Los Alamos National Laboratory and earlier pellet production at the Mound Facility. The SRS pellet production process, starting with calcined plutonium oxalate powder, consisted of the following steps:

1. Oxygen exchange to reduce neutron radiation by exchanging the naturally occurring ^{17}O and ^{18}O with ^{16}O by heating 5 hours at 800°C in ^{16}O.
2. Outgas for 1 hour at 1000°C in ^{16}O to free stored decay helium from the powder before particle sizing steps are initiated.
3. Ball mill for 12 hours at 100 rpm to produce a more uniform shape and an average particle size of 1.4 mm.
4. Granule formation by compacting the milled powder at ambient temperature to about 50% of theoretical density, sizing to <125 mm by forcing (with a roller) the material through a sieve.
5. Sinter 40% of the material at 1600°C and 60% at 1100°C for 6 hours in argon to densify the granules.
6. Sieve the densified granules to eliminate agglomerates that may form due to the self-heating of ^{238}Pu.
7. Blend the sintered and sieved material.
8. Vacuum hot-press at 1525°C and 19.4 MPa in inductively heated graphite dies to form the pellets.
9. Heat treat for 6 hours at 1525°C in an oxidizing atmosphere to assure dimensional stability, reoxidize to a O/Pu ratio of 2.00, and remove volatile impurities.
10. Vacuum outgas at 1500°C for 1 hour to reduce detrimental impurities and reduce the O/Pu ratio to 1.99 and thereby reduce material transport within the final capsule.

These ten process steps produced a chemically and dimensionally stable pellet that had the desired density, O/Pu ratio, porosity distribution, and other quality measures.

Encapsulation of Fuel Forms

Primary containment for each radioactive pellet is provided by a shell of iridium alloy (Kanne 1983). Iridium is a platinum-group metal that is compatible with plutonium oxide and has good strength and impact resistance at the 1310°C heat source operating temperature. Iridium is unusual in its inertness, high density, and rapid work hardening.

The containment capsules were thin-wall cylindrical shells that were 0.025-inch thick. A vent assembly in the end of the capsules allows the escape of helium gas from radioactive decay, but it does not allow escape of PuO_2 fines that may develop during the service life of the pellet. The welded butt joint was backed by a 0.005-inch thick foil of iridium to minimize the effect of weld heat on the PuO_2 pellet. A unique shipping container was designed at SRS for shipping the welded capsules to the Mound Facility for assembly into an RTG. The capsules were held in graphite felt within the shipping container. At the Mound Facility, the capsules were loaded into a graphite matrix before final assembly into the heat source modules.

A gas tungsten arc welding process was used to weld the girth of the iridium alloy capsules. The welding process was adapted from the Multi-Hundred Watt heat source welding process initially developed at the Mound Facility and briefly used at SRS prior to the General Purpose Heat Source program. The welding process produced a full penetration weld by welding at a relatively high speed of 30 ipm. Magnetic arc oscillation was used to promote the desired grain structure within the weld.

Welding the heat source capsules required a unique welding station for operation in the hot cell lines in Building 235-F. The welding equipment was computer controlled at a time when desktop computers were in their infancy. The welding station consisted of a turntable, an upper dead-weight-loaded positioner, and a horizontally mounted torch. Stepping motors actuated the three components. Welding current was supplied by a power source with a dual schedule programmer to accommodate tack welds and full welds. The computer automatically rotated the capsule through a series of three short tack welds and then the full closure weld. The flexibility of the computer-controlled welding equipment was a significant aid in developing the welding process and in creating an efficient production operation.

The welding operation was carried out remotely using manipulator arms and glove ports in the heavily shielded cells in Building 235-F. Remote operation was required because of the toxic nature of the plutionium-238 alpha-emitting radioisotope that was encapsulated. Welding speed was a compromise between higher speeds that produced centerline grain boundaries and slower speeds that resulted in through-wall grain boundaries. Welding current was a compromise between high current that caused the formation of a large columnar grain structure and low currents that could lead to lack of penetration. Small grain size with irregular boundaries was desirable. A hot cracking problem resulting from liquation of grain boundaries was minimized by four-pole arc oscillation. Hot cracking was eliminated in production capsules by introducing a state-of-the-art ultrasonic test that was used to cull out capsules with cracks and assured that only high quality welds were placed in service.

Plutonium Fuel Form Facility (PuFF)

The Pu Fuel Forms Facility (PuFF) was constructed in Building 235-F, adjacent to the Neptunium Billet Line. The Billet Line was the source of the Np-Al billets extruded for Mark IV production of Pu-238. The first floor of the PuFF consists of two remote manipulator lines (see Figure 2). The East Line conducts the Pu-238 powder receiving, processing, hot pressing and furnaces, and the West Line contains the welding of iridium cladding, decontamination, and welding of shipping containers. Five gloveboxes are attached to the maintenance side

Figure 2. Pluntonium Fuel Form Facility Hot Cell, Building 235-F

of the East Line, and the vacuum hot press. Also on the first floor is the Pu Experimental Facility, with full-size powder processing capabilities similar to the Puff East Line. An innovative inert gas system provided ventilation for the East Cell Line, as well as emergency and maintenance ventilation. A complete metallography glovebox line on the second level was used to examine pellet production and iridium sample welds. Final facility checkout was completed in 1977, and immediate production began on Multi-Hundred Watt spherical fuel. The facility was then reconfigured for a longer campaign of General Purpose Heat Source pellet production. Technical difficulties were encountered and overcome: rapid failure of glovebox gloves, highly mobile Pu powder, rapid pellet cracking, and underbead cracking of the iridium welds. The PuFF produced General Purpose Heat Source product that was acceptable when tested by high-speed impact at Los Alamos National Laboratory. The PuFF Facility was shut down at the end of the Ulysses fuel production, and the remaining NASA requirements have been met by Los Alamos.

Acknowledgments

This paper is dedicated to D. Thomas Rankin, friend, colleague, valued scientist, and one of the technical leaders throughout the SRS Pu-238 heat source program. Tom died October 23, 1999.

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Biographies

D. Thomas Rankin

Thomas Rankin was an advisory scientist and a program manager of the SRTC plutonium immobilization program at the time of his death in October 1999. Early in his career, he worked on the use of various isotopes for heat sources applications before playing a major role in the application of Pu-238 for the General Purpose Heat Source program. He received his B.S. in 1963 and his Ph.D. in 1967 in ceramics from Rutgers University. He was with SRTC since his graduation, except for duty as a military commander during 1968-70 at the U.S. Army Materials & Mechanics Research Center. He was a fellow and member of the Board of Trustees of the American Ceramics Society.

William R. Kanne, Jr.

William Kanne is a senior fellow engineer at the Savannah River Technology Center. He worked on encapsulation of MHW and GPHS heat sources during the late 1970s and 1980s. He has 30 publications and one patent on topics that include welding of irradiated materials, corrosion, and resistance welding in addition to welding of heat source capsules. Mr. Kanne is a graduate of the Johns Hopkins University with a B.A. in physics. He received a Ph.D. in metallurgical engineering from the University of Wisconsin in 1968, and he has been at Savannah River Site since that time. He is currently on the Board of Directors of the International Metallographic Society and is active with the American Welding Society, ASM International, TMS, and ANS.

McIntyre R. Louthan, Jr.

McIntyre Louthan is a senior advisory engineer in the Materials Technology Section of Savannah River Technology Center. He is the author of approximately 200 technical publications, editor of nine books and a fellow in ASM International. Mr. Louthan developed the lecture "Why Stuff Falls Apart", which has been given over 200 times to colleges, universities, and civic and professional organizations. He has served as president of the International Metallographic Society, a member of the Board of Trustees of the National Youth Science Foundation, and chairman or co-chairman of 12 international conferences. He was a key reader for Metallurgical Transactions, a member of the editorial advisory board for Materials Characterization, and the series editor of Microstructural Science. Mac has given invited presentations throughout the U.S., Canada, Europe, and Asia and is a member of Sigma Xi, Alpha Sigma Mu, and Tau Beta Pi.

Dennis F. Bickford

Dennis Bickford is an advisory engineer at the Savannah River Technology Center where he is principally concerned with melter design and glass melting for DWPF and Idaho National Environmental Engineering Lab. Denny was Separations Technology manager for Building 235-F Operations during the startup of the Plutonium Fuel Form Facility and during production of the fuel for the Galileo and Ulysses missions. He received a B.S.M.E and B.S. in Metallurgy and Materials Science from the Massachusetts Institute of Technology and a M.S. in Metallurgy and Materials Science from Carnegie-Mellon University. Postgraduate studies include Ceramics Engineering at Clemson University. He is a member of Pi Tau Sigma mechanical engineering honorary society and Phi Kappa Phi. He is a fellow of the American Ceramic Society and president of the Board of the National Science Foundation Center for Glass Research.

James W. Congdon

James Congdon is a senior fellow scientist at the Savannah River Technology Center. He has worked at SRTC since 1977 in material and process development. He assisted in the development of the fabrication process for the General Purpose Heat Source and later developed several process improvements. Mr. Congdon is currently leading the SRTC effort to develop a process to immobilize excess plutonium. He received his B.S. in 1973 and his Ph.D. in 1978, both from Alfred University in Ceramic Science. He is a member of the American Ceramic Society.