Reactor Production Diversity

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Abstract

Production diversity of Savannah River reactors was amply demonstrated by their efficiently producing of over 100 exotic radioisotopes, even though the original design mission was primarily to produce plutonium and tritium. Production versatility of Savannah River Site (SRS) reactors is unparalleled as a consequence of high power, large range of neutron flux, excellent neutron economy, and large capacity for a variety of target materials.

Operation at high neutron flux was perhaps the most outstanding advance in reactor technology made at SRS. Operation at neutron flux levels comparable to neutron flux levels of reactors specifically designed for high flux operation made SRS the principal source of synthetic nuclides for the free world from 1965 through 1970.

Records achieved during the High Flux Demonstration include a thermal neutron flux level of 6.1×10^{15} neutrons per square centimeter per second, a thermal heat flux level of 2.66×10^{6} BTU per hour per square foot, and a fuel power of 226 kW per gram of uranium-235. An even higher neutron flux level of 7×10^{15} neutrons per square centimeter per second was achieved later during the Californium Production Campaign. This record neutron flux level was achieved during the time when the High Flux Isotope Reactor (HFIR) was being built at Oak Ridge, Tennessee. The peak design neutron flux level for the HFIR was 3-5 $\times 10^{15}$ neutrons per square centimeter per second; a value less than that achieved in SRS reactors.

Theoretically, an SRS reactor is capable of operation at high neutron flux level while supporting up to 130 kilograms of target material compared to a maximum of 0.3 kilograms for the High Flux Isotope Reactor. However, only nine kilograms of target material was available for irradiation during the Californium-I Production Campaign.

Several world production records were achieved during High Flux Operation:

- The most californium-252 ever made (2.1 g).
- The highest specific activity cobalt-60 ever produced (700 curies per gram)
- The most polonium-210 ever made (over 0.5 kilogram)

Additional information about these and other SRS major reactor products is summarized in Table 1.

Background

Savannah River Site (SRS) reactors are believed to be the most versatile nuclear reactors ever operated. They are large, comparable in size and power level to commercial power reactors; but unlike commercial power reactors, they have been operated at neutron flux levels as high as those achieved in research reactors specifically designed for that purpose. They have proved to be extremely versatile by their ability to simultaneously produce a wide variety of radioisotopes. Over 100 different radioisotopes were produced in SRS reactors between 1954 and shutdown in 1988.

SRS reactors use heavy water (D_2O) for both coolant and moderator. This design feature contributes considerably to reactor versatility and the ability to achieve very high neutron flux levels because heavy water absorbs very few neutrons and is also an excellent media for slowing down neutrons so they can be captured in target materials.

Co-production of electricity and radioisotopes was briefly considered during conceptual design, but the idea was soon abandoned for two reasons:

- Long operating cycles are essential for efficient generation of electricity, whereas relatively short cycles are required for production of weapons-grade plutonium.
- High reactor coolant temperature is required to provide steam for turbine generators to generate electricity. High coolant temperature is incompatible with aluminum cladding. Aluminum cladding is extremely desirable because it is inexpensive, absorbs very few neutrons, and is an outstanding barrier to contain tritium in lithium-bearing targets and control rods.

SRS reactors were originally designed to operate at about 400 megawatts thermal (MWt). By 1964, the reactors were routinely operated at powers over 2400 MW_t during the winter months. In 1967, C Reactor achieved a peak power of 2915 MW_t, the highest power ever achieved in an SRS reactor. This reactor power increase of over a factor of six represents one measure of the extreme versatility that has become a hallmark of SRS reactors.

Both upgrading the reactor hydraulic system and developing sophisticated fuel and target assemblies contributed to the ability to operate at high power levels. Physical modifications required to increase reactor power are described in the article, "Increased Production," by James M. Morrison. Sophisticated fuel and target assemblies were designed and developed at the Savannah River Laboratory to take advantage of the higher power potential of the improved reactor cooling system. Development of these assemblies is described in articles by Philip Permar and William McDonell.

In his January 1964 State of the Union message, President Lyndon Johnson ordered the shut down of several weapons production reactors due to a decreased requirement for weapons materials. As a consequence of the demonstrated versatility of SRS reactors, only R Reactor was shut down as compared to the shutdown of several Hanford reactors. This trend continued until only one Hanford reactor (N Reactor) remained on-line, while three SRS reactors continued to operate. The reason for keeping SRS reactors in service was their demonstrated capability to operate over a wide range of neutron flux levels while simultaneously producing a variety of radioisotopes.

The only reason that N Reactor continued to operate was because it co-produced about 800 MW of electricity for the Northwest. It was incapable of efficient production of both electricity and weapons-grade plutonium. Relatively short reactor cycles are required to make weapons-grade plutonium whereas long fuel cycles are necessary for efficient production of electricity. This is the very reason why co-production was ruled out during conceptual design of SRS reactors.

Reactor Description

Each Savannah River Site (SRS) reactor has approximately 600 4-inch-diameter positions that may be used for either fuel or target assemblies. Fuel assemblies normally contain U-235-bearing material while target assemblies contain the material to be transmuted into the desired radioisotope. The reactor core is the combination of all of the fuel and target assemblies. Fuel and target material can either be contained in the same assembly or in separate assemblies. A reactor core of fuel and target in the same assembly is referred to as a uniform lattice; whereas, if these materials are in separate assemblies, the core is called a mixed lattice. One of the major keys to SRS reactor versatility is the ability to simultaneously produce many different radioisotopes in a mixed-lattice core. Fuel and target assemblies are suspended vertically from an inlet process water plenum. Deuterium oxide (D_2O) is used for both coolant (dissipates fission heat) and moderator (slows down neutrons so they can be absorbed in fuel or target materials). Neutron loss due to absorption in D_20 is very low

compared to other moderating materials, such as graphite or light water (H_2O). The so-called "neutron economy" is much better with D_20 moderator than with other moderating materials.

The reactor core is enclosed in a cylindrical stainless steel tank that is about 15 feet tall and 16 feet in diameter. Process water (D_20) flows down from the inlet plenum through the fuel and target assemblies to remove heat, and is then discharged into the space between the fuel and target assemblies before exiting the reactor tank. D_20 coolant exits through 6 effluent nozzles at the bottom of the reactor and is then pumped through 12 heat exchangers before return to the reactor plenum. Light water coolant flowing through the shell side of the heat exchanger dissipates fission heat to a cooling pond (R, P, and L reactors), a cooling tower (K Reactor), or a stream (C Reactor) that flows into the Savannah River.

Core Configurations

SRS reactors were originally designed and built to produce plutonium and tritium for nuclear weapons. The first reactor core consisted of four columns of aluminum-clad natural uranium slugs in each of the 600 reactor lattice positions. Reactor power was controlled by lithium-bearing control rods that produced tritium. All future reactor configurations employed lithium-bearing control rods, except those designed for high flux. Because lithium melts at a relatively low temperature, cadmium or cobalt was used in control rods for reactor cores that operated at elevated neutron flux levels, and, hence, high fuel specific power (MW_t per linear foot of fuel).

The crux of a nuclear reactor is the core design. Commercial power reactors use essentially the same basic core design for the life of the reactor. The U-235 content of fuel assemblies may vary somewhat, but the overall configuration of the core stays the same. By contrast, almost two dozen distinct reactor cores were designed, developed, and operated in Savannah River reactors. Furthermore, several additional cores were designed and developed, but were discarded for various technical reasons. Each core design is analogous to a different reactor because each design required developing fuel and target assemblies and nuclear physics experiments in test reactors to confirm the physics before irradiation in a production reactor. Each new core design required a complete safety analysis to confirm that it would perform within acceptable limits even under adverse conditions. Advanced reactor core designs were conceived and developed by engineers and metallurgists in the Savannah River Laboratory.

Almost half of the reactor cores were designed to produce plutonium for weapons. Uniform-lattice core designs consisted of either solid slugs of uranium, hollow slugs, or nested tubes. Hollow slug and nested tube designs were developed to increase both flow area and heat transfer surface area. All of the slug designs used natural uranium (0.71% U-235). Nested tubular designs ranged from natural uranium to 1.1 wt % U-235. More flow and heat transfer area contributed to higher reactor power while higher U-235 content contributed to increased amount of Pu-239 produced per megawatt day.

High Flux Demonstration

The crowning technical achievement of SRS reactors was operation at high thermal neutron flux in C and K Reactors during the mid 1960s. In 1961, President John F. Kennedy appointed Dr. Glenn T. Seaborg as chairman of the Atomic Energy Commission. Prior to that appointment, Dr. Seaborg shared the 1951 Nobel Prize for chemistry with Edwin M. McMillan for the discovery of artificial elements heavier than uranium, including plutonium, americium, curium, berkelium, and californium. Naturally, Dr. Seaborg had a strong interest in production of sufficient quantities of these artificial elements for scientific evaluation, which might lead to a commercial market once their properties were fully known. Hence, Dr. Seaborg was instrumental in directing the Savannah River Operations Office to authorize high flux opera-

tion in a Savannah River reactor to produce gram quantities of californium and other transuranium isotopes.

In the same time frame, a high flux isotope reactor (HFIR) was designed and built at Oak Ridge to produce small quantities of artificial nuclides for scientific evaluation. Operation of an SRS reactor before completion of the HFIR would provide an opportunity to produce target material (plutonium, americium, and curium) for the HFIR and also to evaluate the reactor performance of target assemblies that would later be used in the HFIR.

Radioisotopes that require multiple neutron captures and that have short half-lives can only be made in quantity by irradiating target material at high neutron flux levels. Neutron flux level is directly proportional to fuel specific power (megawatts per foot of fuel assembly) and inversely proportional to fuel U-235 content (grams of U-235 per foot of fuel assembly). The most efficient production of plutonium and tritium is achieved at a thermal neutron flux level of about 7 − 10 x 10^{13} neutrons per square centimeter per second. By contrast, optimum production of Cf-252 requires a hundredfold higher neutron flux level because of the multiple neutron captures required to produce it, and the fact that its half-life is only about 2.5 years.

A decision was made to demonstrate high flux operation in C Reactor prior to the actual production campaign in K Reactor. To achieve high flux, the number of fuel positions was decreased from approximately 600 to 107, the active length of U-235 in the fuel assemblies was reduced from about 12 to 6 feet, and the fuel loading was reduced to 25 grams of U-235 per foot. This combination of changes resulted in increasing the neutron flux level from about 7 x 10^{13} to 6 x 10^{15} neutrons per square centimeter per second.

Target material consisted primarily of Pu-242. In addition, three 1-inch-diameter thimbles contained Am-243, Cu-244, and 150 nuclides of 66 elements for 9 universities and laboratories.

Control rods for the high flux demonstration contained cadmium, which could withstand operation at higher temperature without melting the lithium, which is normally used to produce tritium in low flux operation. Cadmium in control rods was replaced with cobalt in the following Californium Production Campaign.

The High Flux Demonstration began in February 1965 and lasted a year. A neutron flux level of 6 x 10^{15} neutrons per square centimeter per second was achieved, and the demonstration was considered a success by paving the way for the following Californium Production Campaign.

Production of Radioisotopes

In addition to the relatively large-scale production of radioisotopes described in the following sections, small amounts of 150 different target materials were irradiated primarily during high flux operation. Irradiation of these samples was requested for research by Argonne National Laboratory, Oak Ridge National Laboratory, and the University of California laboratories at Berkeley and Livermore. A list of samples irradiated during the High Flux Demonstration is contained in DP-999, "The Savannah River High Flux Demonstration."

Californium-252

Based on technical information gained through operation of the High Flux Demonstration in C Reactor, an improved reactor core was developed to produce Cf-252 in K Reactor, which began operation in August 1969. The improved core consisted of 96 large-diameter assemblies, each of which contained 3 concentric tubes of U-235 alloy fuel rather than the 2-tube assemblies used for the High Flux Demonstration. Eighty-six of the fuel assemblies contained Pu-242 in a four-foot section of the outer housings. Irradiated fuel was discharged separately from the outer housing, which remained in the

reactor until termination of the production campaign. The reactor core also contained six assemblies of Am-243 and Cm-244 target material. Each fuel cycle lasted 3.5 to 4 days, and was followed by a 12 to 18 hour reactor shutdown to replace irradiated fuel tubes.

The reactor core of fuel assemblies was surrounded by 104 target assemblies containing Np-237 to produce very pure Pu-238. These assemblies were located in the reflector between the core and the reactor tank wall.

A peak flux of 7 x 10^{15} was achieved, and 2.1 grams of Cf-252 were produced before termination of the production campaign on November 9, 1970, due to failure of an antimony-beryllium source rod, while the reactor was being refueled. A second Californium Production Campaign was planned, but it was not executed because the market for Cf-252 never materialized as originally expected.

In 1965, Carl Schlea and Dean Stoddard of the Savannah River Laboratory proposed using Cf-252 in needles for cancer therapy. The key to this proposal is the idea of treating localized cancers with neutrons from Cf-252 encapsulated in needles similar to those used to contain radium. The problem with radium needles inserted in a tumor is that radium emits X-rays, which do not discriminate between cancerous cells and healthy cells; hence, treatment can be taken only with the risk of destroying healthy tissue. Basing their research on theories previously developed, Schlea and Stoddard concluded that Cf-252 might provide a lethal dose of radiation to a tumor without irreparable damage to adjacent normal cells. They recognized that treatment would be most effective for tumors located too close to vital organs to be removed surgically.

As a consequence of the Schlea-Stoddard proposal, a Cf-252 needle manufacturing facility was designed and installed in the Savannah River Laboratory. Over 3000 needles were made that contained from 5 to 140 micrograms of Cf-252 for evaluation by numerous hospitals and universities here and abroad.

Tritium

The Savannah River Plant was built primarily to produce tritium, an essential ingredient in thermal-nuclear weapons. Tritium significantly increases the yield of nuclear weapons. Over the operating life of the reactors, six different reactor cores were designed, developed, and implemented to produce tritium. All of the tritium-producing cores were of uniform-lattice design. That is, both the fuel (U-235) and the target material (lithium) were contained in the same assembly, but in separate slugs or tubes within the assembly.

The Mark 22 was the most efficient of the tritium-producing cores because it was designed to use the full capacity of the reactor hydraulic system and was compatible with the use of outer aluminum housings that could be recycled for several years of operation. In previous designs, the outer housing contained lithium and was discharged from the reactor with the rest of the assembly each time the core attained its goal exposure (megawatt days).

One tritium-producing core (Mark VI-E) was designed with a removable inner lithium target so that the fuel could be irradiated to a very high exposure. The high exposure depletes about 70% of the U-235 and thus concentrates U-236, which is formed from every sixth neutron capture in U-235. Irradiation of U-236 forms neptunium-237 (Np-237), which is the target material for production of Pu-238. Hundreds of kilograms of tritium were produced during the 34-year operating life of the SRS reactors.

Plutonium-239, -240, and -242

Plutonium for weapons was produced in one or more SRS reactors every year from initial operation in 1954 through the final shutdown in 1988. Approximately half of the two dozen

reactor cores that were operated in SRS reactors were designed primarily to produce plutonium. The initial cores contained the fissile isotope (U-235) and the target isotope (U-238) in the same assemblies (uniform lattice). Mixed-lattice reactor cores to produce plutonium began operation in 1968. The mixed-lattice core consisted of an equal number of highly enriched uranium (U-235) fuel assemblies and depleted uranium (U-238) target assemblies. The fuel assemblies supplied the neutrons, and the product (Pu-239) was produced in the target assemblies.

The most productive plutonium-producing reactor core (Mark 15) was a uniform lattice of slightly enriched uranium (1.1 wt % U-235) in assemblies of concentric tubular slugs. The outermost 84 reactor positions contained lithium-bearing tubular assemblies, which protected the reactor tank wall from excessive neutron damage and reduced argon formation between the reactor tank and the surrounding thermal shield.

Plutonium was produced at assays that ranged from 3% to about 40% Pu-240. Weapons-grade plutonium is defined by both the Pu-240 and Pu-241 assays. The very high assay Pu-240 material was produced for use as target material for production of higher radioisotopes and for breeder reactor studies that use plutonium as fuel. Hundreds of kilograms of Pu-239 and Pu-240 were produced between 1958 and 1984.

Tens of kilograms of Pu-242 were produced by irradiating large quantities of Pu-239 over a period of several years.

Plutonium-238

Plutonium-238 generates 0.5 watt per gram when it decays by spontaneous emission of alpha particles, which are easily blocked by a thin metal sheath. These properties coupled with a relatively long half-life (about 89 years) make Pu-238 an ideal heat source for use in thermoelectric generators for space exploration. SRS began production of Pu-238 by the irradiation of Np-237, a byproduct of the fission of U-235. Experiments left on the moon by "Apollo" crews were powered by Pu-238 made at SRS. Unlike solar cells, generators powered by Pu-238 are ideal for exploration of the outer planets.

The challenge in making Pu-238 is to minimize formation of Pu-236, which is formed by absorption of fast neutrons. The solution to this problem was design of reactor lattice configurations that maintain adequate separation of fast neutrons from neptunium target material. To accomplish this separation, neptunium targets were located in the D_2O reflector beyond the fuel in high flux cores.

By 1978, over 300 kilograms of Pu-238 had been made in SRS reactors.

Another route to formation of Pu-238 that avoids the offensive Pu-236 isotope is producing it as a decay product of Cm-244. In 1970, Pu-238 was separated containing less than 0.3 parts per million of Pu-236

Curium-244

Curium-244 is produced through successive neutron captures in Pu-239. Curium -244, like Pu-238, decays by alpha emission but produces five times as much heat (2.5 watts per gram) as Pu-238. However, the half-life of Cm-244 is only about 18 years compared to about 89 years for Pu-238; hence, Pu-238 is the preferred isotope for long-term space missions.

Gram quantities of Cm-244 were first produced in SRS reactors in 1962. In 1964, a special reactor core began irradiation to produce kilogram quantities of Cm-244 for evaluation as an alternative to Pu-238 and for use as target material to produce Cf-252. The reactor core for this production campaign (called Curium I) was designed to operate at a higher neutron flux level than previous SRS reactor cores. The targets from this irradiation were reprocessed

and made into new targets for the second phase of this program (called Curium II). By 1967, a total of 5.9 kilograms of Cm-244 had been made during the two curium production campaigns.

Continued irradiation of plutonium target material eventually produced about 12 kilograms of Cm-244.

Uranium-233

A breeder reactor produces more fissionable material than it consumes to maintain a nuclear chain reaction. Uranium-233 (U-233) is such a material when used in conjunction with thorium targets. Uranium-233 is produced by neutron capture in thorium, which is an abundant element in nature.

Admiral Hyman G. Rickover was a key figure in the development of the U.S. nuclear navy and commercial reactors. One of his projects included developing a breeder reactor using the Shippingport Reactor located in Pennsylvania. SRS was requested to produce several hundred kilograms of U-233 for Admiral Rickover's program.

By 1956, U-233 was being produced in small quantities, but it was contaminated with U-232, which emits strong gamma radiation and, hence, made fuel fabrication from this material very difficult. In the original irradiations, the fissionable U-235 and the thorium targets were in the same assemblies. Since U-232 is produced from interaction with fast neutrons, the challenge was to separate the fissionable U-235 from the thorium target material. This separation of fuel and target material allowed the neutrons reaching the thorium to be slowed down by the D_2O moderator, thus greatly reducing the formation of U-232.

By 1965, U-233 was being produced in quantity with only 3 - 6 parts per million of U-232. Several hundred kilograms of U-233 were produced through 1968.

Polonium-210

Polonium-210 is made by irradiating targets of bismuth-209. Operation at high neutron flux with small reactor cores during the 1960s provided an opportunity to demonstrate the production of large quantities of Po-210 by irradiating bismuth targets in the D_2O reflector between the reactor core and tank wall. Approximately 0.6 kilogram of Po-210 was made that way.

Use of Po-210 in thermoelectric generators was briefly considered, but this application was abandoned because of its short half-life (130 days) and difficulty in separating polonium from bismuth.

Cobalt-60

Irradiation of Co-59 produces Co-60, which emits very penetrating gamma radiation. Sources made of Co-60 can be used for cancer treatment, food sterilization, radiography, and as heat sources for thermoelectric generators.

Small amounts of low-specific-activity Co-60 (~ 50 curies per gram) were produced in SRS reactors as early as 1955. High flux operation, beginning in 1965, provided an opportunity to make large quantities of very-high-specific-activity Co-60 because Co-59 was the preferred material for control and safety rods in high flux reactor cores. Lithium,

normally used in control rods, melts at a considerably lower temperature than cobalt. Therefore, for reactor safety reasons, Co-59 was substituted for lithium during high flux operation, where a Co-60 specific activity of 700 curies per gram was achieved (the highest specific activity Co-60 ever made). Specific activity of pure Co-60 is about 1140 curies per gram.

Perhaps the greatest interest in Co-60 was manifested by the food industry, which used gamma radiation to sterilize food for long-term storage without refrigeration.

Table 1. SRS Reactor Major Products

Radioisotope	When Produced	Amount	Application
Plutonium-239	1954 - 1988	1000's kg	Nuclear Weapons
Tritium	1954 - 1988	100's kg	Nuclear Weapons
Uranium-233	1956 - 1968	100's kg	Breeder Reactor Development
Plutonium-238	1959 - 1988	100's kg	Thermoelectric Generators for Space Exploration
Plutonium-240	1958 - 1984	100's kg	Target Material for Transplutonium Isotopes
Plutonium-242	1964 - 1984	10's kg	
Cobalt-60	1956 - 1970	~ 66 mega curies	Gamma Radiation Source Heat Generation Sources
Curium-244	1962 - 1978	~ 12 kg	Thermal Electric Generators Target for Production of Transplutonium Isotopes
Polonium-210	1966 - 1969	~ 600 g	Intense Radiation Source
Californium-252	1965 - 1970	2.1 g	Cancer Treatment, Oil-Well Logging, etc.

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Definitions

Neutron Flux - Neutrons per square centimeter per second

Fuel Specific Power - Megawatts of thermal heat output per linear foot of fuel

Half-life - The time required for half of a radioisotope to disappear through radioactive decay

Target Material - Material to be irradiated with neutrons for transmutation to different radio-isotopes

Pure Plutonium-238 - Contains less than 0.3 ppm of plutonium-236, which is a contaminant due to its gamma radiation

SRS - Savannah River Site.

MW_t - Megawatt thermal (heat produced by nuclear fission)

Co-production - The simultaneous production of nuclear materials and generation of electricity

Uniform Lattice - A reactor core in which all of the assemblies are alike

Mixed Lattice - A reactor core containing a mixture of uranium-235-bearing assemblies and target assemblies

Neutron Economy - Fraction of useful neutrons

Biography

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1953 - Savannah River Plant, Works Technical Department; Engineer, Heavy Water Technology; Engineer, Reactor Technology

1959 - Process Supervisor, Reactor Technology

1962 - Senior Supervisor, Reactor Technology

1963 - Savannah River Laboratory; Nuclear Material Division; Reactor Engineering Division

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