USE OF LEU IN THE AQUEOUS HOMOGENEOUS MEDICAL ISOTOPE PRODUCTION REACTOR

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ABSTRACT

The Medical Isotope Production Reactor (MIPR) is an aqueous solution of uranyl nitrate in water, contained in an aluminum cylinder immersed in a large pool of water which can provide both shielding and a medium for heat exchange. The control rods are inserted at the top through re-entrant thimbles. Provision is made to remove radiolytic gases and recombine emitted hydrogen and oxygen. Small quantities of the solution can be continuously extracted and replaced after passing through selective ion exchange columns, which are used to extract the desired products (fission products), e.g. molybdenum-99. This reactor type is known for its large negative temperature coefficient, the small amount of fuel required for criticality, and the ease of control. Calculation using TWODANT show that a 20% U-235 enriched system, water reflected can be critical with 73 liters of solution.

Introduction

The need in nuclear medicine for the fission product isotope, molybdenum-99, is well known and has been the subject of investigations and presentations at previous RERTR conferences. The concentration of effort has been on creation and processing of LEU targets which are subsequently irradiated in high flux reactors. Other papers in this Session describe further efforts along this line.

The Medical Isotope Production Reactor (MIPR) is a design under development by Babcock & Wilcox (B&W) which eliminates the need for target preparation and dissolution. It is particularly suited for the use of LEU in medical isotope production. The MIPR is an aqueous homogeneous reactor operating at 200 KW thermal power and produces over 2000 curies of Mo-99 per DAY.
Reactor Description

The reactor is composed of a solution of uranyl nitrate (UO$_2$(NO$_3$)$_2$) in water. The reactor can be designed to function with U-235 enrichments of 18% to 90%. One variant of the MIPR design, using LEU (uranium enriched to 20% U-235), has been analyzed at Los Alamos National Laboratory by Dr. Robert Kimpland. Using TWODANT with an Sn of 16 to model the core, the uranium concentration is 150 grams/liter and the core volume is 73 liters, giving a critical height of 460 mm. The core would contain about 2.2 kg of U-235 or about 18 kg of salt. The container was assumed to be stainless steel and is fully reflected by water. A second variant used an aluminum container and 100 liters of solution. The critical concentration was 117 gram of uranium/liter, 20% enriched. This means the U-235 content of the whole reactor is 2.34 kg.

In the reference MIPR system, the core container is assumed to be aluminum. The container is cylindrical with cooling fins. On the top cover are stepped aluminum thimbles which permit the insertion of control and safety rods and small tubing for solution addition and removal. The solution is acidic to prevent corrosion of container and thimbles. The entire reactor container is immersed in a pool of water assumed to be 3 by 3 meters and 7 meters high (63 m$^3$, or 63 metric tons).

The burnup of the fissionable material, U$^{235}$, is about 1.2 grams per megawatt-day. For a 200 kWt operation, the burnup is less than one quarter gram per full power day. Additions to the reactor core to compensate for burnup can be through the addition of a few grams of solution per month.

The planned power level of the reactor is 200 kWth and the operating temperature of the uranium solution is 80°C (176°F).

The proposed reactor solution container is a cylinder with a volume of 100 liters, approximately 450 mm diameter and 750 mm height. The outer and inner surfaces are finned for enhanced heat transfer. The material is an aluminum alloy chosen for strength and corrosion resistance. The actual size and composition of the container, however, will depend on confirmatory thermal/hydraulic, neutronic, and materials performance evaluations. Figure 1, "MIPR System Design", shows the reactor at the bottom of a pool of ordinary water. The various gas recombiners, filters and traps are under water. The extraction columns are also under water and are fitted with "quick disconnect" fittings for easy replacement. The valving is arranged so that a column can have a small amount of reactor solution pasing through for molybdenum stripping. The valving also permits wash and eluting solutions to pass through the column for the cleaning and removal of the product. Heat removal from the fuel solution container is passive through extended surfaces on the container and is absorbed by the bulk pool water. The pool water is cooled by passing it through a conventional tube-shell heat exchanger. The normal pool water temperature is 22 °C. The water on the secondary side of the heat exchanger is cooled using a forced draft cooling tower.
2. Gas Handling

During operation of the MIPR, two types of gas are produced. The radiolytic dissociation of the water results in the production of hydrogen and oxygen. The fission process produces some fission products which are gaseous at standard temperatures and pressures.

Extrapolating from the experience of the SUPO reactor at Los Alamos (RH 13-129), hydrogen gas evolution (and by inference the oxygen evolution, half mole of oxygen for each mole of hydrogen) for a 200 kWt reactor would produce 600 cc/sec of H₂ or 60 milligrams/sec. However, as in the case of pressurized water reactors, it may be possible to operate the reactor with a hydrogen overpressure (about 50 psig) which may obviate the need for a recombiner, or, as in the case of the Homogeneous Reactor Experiment at Oak Ridge, internal recombinaton can be possible with a copper catalyst.

Approximately 60% of the fission products have a xenon or krypton isotope as members of the decay chains. Half of the isotopes, about 15 cc/day for 200 kWt, are stable or have half-lives that permit them to be removed from a fluid fuel before they decay (RH 13-71).

Catalytic recombiners for hydrogen and oxygen have been used industrially for many years. Systems with platinum and palladium catalysts deposited in concentrations of 0.03 to 0.3 weight percent on small (1/8 inch) cylinders of alumina have proven effective for recombining hydrogen and oxygen in non-combustible mixtures. Adsorption of the
Krypton and xenon on activated carbon or other adsorbents is a convenient method of storing the gases until most of the radioactivity decays and can be discharged. Most radioactive containers of charcoal can be stored underwater in the pool until essentially all gases except the 10.3-year Krypton-85 have decayed. The production rate of Kr-85 is 110 millicuries per 200 kWt-day.

3. Instrumentation & Control

The excess reactivity of the MIPR is deliberately kept small to reduce the energy of any postulated incident involving reactivity insertion. This excess reactivity and safety shutdown for deep sub-criticality is achieved by the insertion of solid neutron absorbers, such as clad boron carbide, through re-entrant thimbles. Two safety rods are held by electromagnets which are driven from a motor drive. A control rod is similarly placed but without the electro-magnet.

Void and temperature coefficients of this type reactor have been extensively studied and are large and negative. The concentration of U\(^{235}\) in the solution is selected such that at operating conditions with movable control and safety rods out of the core, the reactor operates near the design temperature and full power. Fission product poisons and burnup of the U\(^{235}\) are periodically compensated for by either control rod movement or the addition of a small amount of U\(^{235}\). The MIPR is configured at maximum reactivity so that geometric changes reduce reactivity and power.

The neutron detectors, source and other radiation detectors can be placed in thimbles which are around the main reactor core tank. Temperature sensors, such as thermocouples or resistance thermometers can be placed in the solution in sheaths or thimbles. Pressures, flows, pump indications, fans, motors, and other parameters which are required to be sensed, conditioned, and displayed or stored are expected to be of conventional design.

4. Heat Removal and Transport

The previous designs of aqueous homogeneous reactors have used internal cooling coils to remove the heat from the reactor solution. The largest power removed was 50 kWt from the ARF reactor in Chicago, Illinois and the KEWB reactor at Atomics International in Canoga Park, California. The KEWB used 90 feet of stainless steel tubing, \(1/4\) inch O.D. with inlet water at 85°F and outlet 109°F. The average fuel temperature was 176°F. The area of the tubing is about 5500 cm\(^2\). The vertical wall area of the MIPR tank is 10,000 cm\(^2\) and the equivalent area can be more than doubled with fins. Avoiding the use of cooling coils will significantly enhance simplicity, reliability, and safety. (KEWB used internal coils rather than external water for cooling because the experimental purpose required a solid shield.)
One shell-and-tube, single-pass heat exchanger is provided to transfer approximately 200 kWt of reactor heat plus 7 kWt from the recombiner. The pool water enters the heat exchanger at 25°C and returns at 22°C. The cooling tower water enters the shell side of the heat exchanger at 16°C and returns to the cooling tower at 18°C. The flow rate for both primary and secondary sides is between 20 and 30 kg/sec. The heat exchanger was designed with 12% excess area for margin. The heat exchanger will be fabricated as an ASME Section III-Class 1 Exchanger using the aluminum alloy 6061.

The secondary cooling system extends from the shell side of the pool heat exchanger to the inlet of the cooling tower and from the cooling tower basin to the heat exchanger inlet. Four-inch aluminum piping connects the heat exchanger to the cooling tower.

A centrifugal pump in the cold leg of the secondary system provides the 28 kg/sec at the 150 meter (500 foot) head to transport the heat from the pool heat exchanger to the cooling tower.

The heat will be rejected to the atmosphere via an induced draft cooling tower. A 15 HP cooling fan draws the air through the tower to remove latent heat from the water. The basin provides 500 ft² of area to retain effluent from the cooling tower.

5. Extraction & Purification of Product

The growth of the Mo-99 in the reactor and in the precipitation column is shown in Figure 2. The extraction flow from the reactor is 1 cc/second. Figure A-2 shows the amount of Mo⁹⁹ in the precipitation column considering that after one day of extraction, the column is valved off (another column would be valved in) and the Mo⁹⁹ decays as it moves through the rest of the process and shipping.

At a maximum 200 kWt operating power, the MIPR can produce 10,000 curies of Mo⁹⁹ per five-day week. Allowing 4 days for processing, packaging, and shipping, and some reactor down time for maintenance/contingencies, the customers would receive approximately 2,100 curies per week.
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