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Title: Robust Medical Isotope Production System

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Robust Medical Isotope Production System

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Introduction

Since 2010 Los Alamos National Laboratory has examined use of aqueous homogeneous reactors (AHR) fueled with a fissile solution of uranium to produce medical isotopes, particularly Molybdenum-99 (Mo-99). Even though AHR have been operated worldwide for seven decades, surprisingly little was known regarding the fundamental physics driving their behavior; therefore, initial efforts were directed at developing a theoretical understanding of AHR physics. This was accomplished through construction of a Dynamic System Simulation (DSS) of AHR systems. These models incorporate families of coupled non-linear differential equations describing the evolution in time of neutronics, thermal-hydraulics, and radiolytic gas generation and transport within the solution core. Models developed were vetted against historical data from SUPO (Super Power), an AHR that operated at Los Alamos from 1951 to 1974, and SILENE, an AHR that operated until 2012 at Valduc, France. SUPO is considered a benchmark for steady-state AHR operation, while SILENE is similarly considered for prompt critical excursions important to safety. Subsequent to verifying the DSS technique on these systems, two other historic AHR, KEWB "A" & "B" (Kinetics Experiment Water Boiler) developed and operated by North American Atomics, were modeled to further demonstrate the validity of DSS for fissile solution systems. The success of this theoretical undertaking provided confidence that the behavior of new and evolving designs of fissile solution systems may be accurately estimated. Scaled up versions of SUPO, subcritical accelerator-driven systems, and other evolutionary designs have been examined. References 1 through 5 present the published results of this body of work.

PROGRAM OBJECTIVES

The theoretical effort described above has provided a basis for examining alternative AHR designs in an attempt to optimize performance as a medical isotope production system. Design objectives match overall Mo-99 production objectives:

- Produce sufficient quantities of Mo-99 to meet national needs
- Development and operational costs consistent with a "full cost recovery" business model
- Design consistent with export control and non-proliferation objectives to provide accessibility of regional facilities in developing countries world-wide

Need for Mo-99

The national need for Mo-99 has been the subject of some debate. Estimates during the 2009 shortage projected quantities based on the number of diagnostic procedures expected to be required by the growing elderly population arising from the "baby-boom" generation. These projections ranged as high as a six percent annual growth rate. The effect of advances in other imaging technologies that tended to suppress the projections were generally not included in these estimates. In addition, the U.S. or even regional North American centric view, failed to consider either global need or global supply chain. Historically, the U.S. domestic supply has been provided principally from Canadian and European producers and the current supply of Mo-99 from Australian and South African producers using Low Enriched Uranium (LEU) targets is growing. The desire of developing nations to provide modern medical imaging technology to their populations is also growing rapidly. The general effect of these trends is to drive the desire for Mo-99 upward.

Cost Considerations

The current global supply of Mo-99 is met by producers that rely on highly subsidized reactors. Without government subsidies Mo-99 would be largely a non-existent commodity. A major contributing factor to this situation is the current market for Mo-99, which is estimated to be in the range of $180M - $200M USD annually. The requirement for "full cost recovery" essentially means that a commercial entity must design, develop, construct, license and subsequently operate a Mo-99 production facility at a profit even with amortization of start-up costs over a reasonable number of initial operating years. Meeting this objective is difficult at best using traditional reactor/target based production, especially when compared to the annual market. Even enjoying 100% market supply by a single producer, initial cost recovery superimposed on annual operating expenses, seems quite illusionary.

An additional constraint that requires careful consideration by any prospective supplier is that medical isotopes are a point-of-use commodity; payment is in units of received Curie. The short Mo-99 half-life of 66 hours results in significant loss of product due to time required for product recovery and shipment.

Associated with the Mo-99 market is the continuing need for other fission based isotopes such as Iodine-131 (I-131) and Xenon-133 (Xe-133). It is worthy of note that those Mo-99 production systems that are being considered that are not based on uranium fission do not address this supply need; hence, the market for these isotopes and the associated opportunity for cost recovery are not realized. Other isotopes, both fission products and potential target produced, can contribute positively to the financial position of any potential producer.

Design Requirements

These considerations suggest a rule-of-thumb domestic need of approximately 10,000 six-day Curies (Ci) for Mo-99 supply. When separation and shipment losses are factored into the need an end-of-radiation (5 days) production quantity of approximately 30,000 Ci may be considered to be a reasonable design target. For a fission-based AHR this equates to a continuous fission power of approximately 1MW over the five day period. Political considerations related to non-proliferation of HEU demand that this value be reached with an LEU-fueled system.

The following requirements have been established for the design of a Mo-99 production system.

1. Produce at least 30,000 Ci of Mo-99 at the end of a 5 day irradiation
2. Maximize the potential for production of other isotopes of medical and industrial interest
3. Utilize fissile solution of Low Enriched Uranium (LEU) as the fuel
4. Cap the cost of facility development to start-up at $150M USD

5. Minimize operating cost by avoiding supply chain for uranium targets
6. Minimize downtime for corrective maintenance

CONCEPT DESIGN

Figure 1 provides a notional design of a system that meets the stated design objectives.

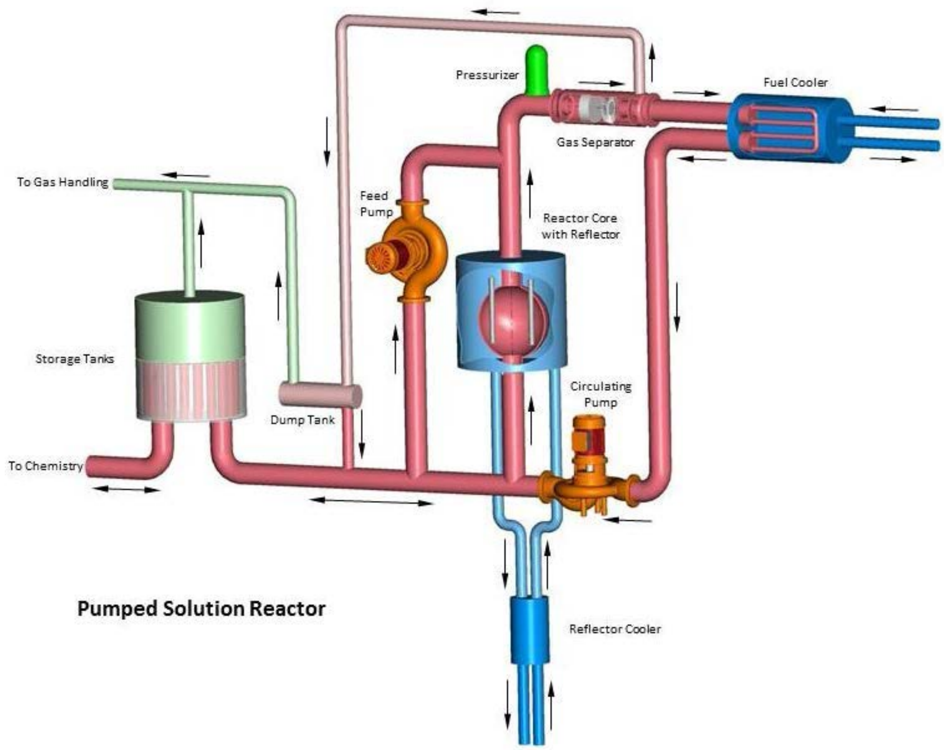

Figure 1: Design Concept for a Robust Isotope Production System

The pumped fuel AHR depicted in Figure 1 utilizes fissile solution fuel of LEU. Key features of the concept include high power operation and overall system simplicity. This is accomplished through circulation of the fuel through a loop, thereby removing the processes of heat transfer and gas separation from the core and performing them elsewhere in the loop. Safety features include multiple reactor shutdown mechanisms and multiple core heat removal pathways. The pumped fuel design takes full advantage of the unique nature of liquid fuels that generally possess docile behavior that lead to very stable operations, particularly at high power. This behavior is caused by two parallel negative reactivity feedback mechanisms, fuel temperature and radiolytic gas void, as well as the large thermal inertia of the fuel itself. Also, fluid fuels can be made to assume critically advantageous shapes or critically safe geometries as desired, and fluid fuel motion facilitates heat removal and gas transport.

The power output of a pumped system is a function of the mass flow rate of the fuel and the excess reactivity of the core configuration. Steady-state power levels in the range of 1-3 MW may be achievable with the concept. The key to this performance is that the thermo-physical behavior of the core is governed by fuel advection. Heat removal from the core occurs by fuel flow carrying away sensible heat. Hot fuel is continuously replaced by cold fuel. Advection of fuel away from the core also continuously sweeps the core of radiolytic gas. Thus, evacuation of gas voids from the core is not dependent upon the buoyancy of the gas bubbles themselves.

The negative reactivity feedback due to fuel temperature rise and void fraction are suppressed because of the thermo-physical effects of fuel advection. This leads to higher power output for a given excess reactivity. Control at high power, may be achieved by altering the flow rate of the fuel, or by adjusting the amount of excess reactivity through changes in control rod position.

The concept possesses several advantages over traditional AHR designs. Because radiolytic gas is transported away from the core in the fuel flow, there is no need for a gas plenum above the core. This eliminates any potential core/plenum interactions that could affect the reactivity of the core. Another major advantage is that heat transfer from the fuel to a coolant does not occur in the core itself. This eliminates the need for cooling structures in the core. No need for in-core plenum or cooling structures means that the core can be simple in geometry and compact, which improves the reactivity worth of the control elements, the control rods and the reflector. Similarly, routine water makeup and other chemical adjustments can be performed outside of the core. Recirculation of the fuel in a loop makes it possible for a wide range of fuel adjustments to occur during operation.

Vessels, piping, and other structures are entirely made of stainless steel. The gas separator, fuel and reflector coolers, pumps and other auxiliary equipment are standard commercial designs. Pressurization is a modest few atmospheres to facilitate fuel flow.

The concept shown utilizes a total fuel inventory of less than 200 liters of LEU solution fuel. It is anticipated that a single fuel load could last as many as 3 years based on operational duty cycle. No replaceable uranium targets are needed since the circulating fuel is the “target”. Separation of product from the fuel could be conducted on a continuous or batch process basis by transferring the desired amount of fuel from the loop to the separation facility, depending on the varying need of customers. Furthermore, continuous processing offers the opportunity for significantly lowering the capacity required of the chemical plant since the full fuel volume would not require separation at a single time.

The estimate of development cost for the AHR is in the range of \$20M-\$25M USD suggesting a complete physical plant including separation chemistry requirements could be reasonably expected to be in the range of \$120M - \$140M USD.

ISOTOPE PRODUCTION

Tables 1 and 2 provide production estimates made at a 1 MW operation, the lower end of estimated power attainable. Table 1 describes the fission product inventory produced in the entire circulating fuel volume that is available for potential extraction. In addition, the proposed design possesses target irradiation cavities within the core, which allow production of various medical and industrial radioisotopes as shown in Table 2.

As can be seen from the tables the pumped system concept can potentially produce a significant quantity of important isotope products. Notice in Table 1 that the Mo-99 levels suggest that a single unit is capable of supplying the entire domestic need of this important medical isotope. The Iodine and Xenon levels allow a similar claim to be made.

Table 1: Fission Products: 5 day operation

Isotope	Symbol	Curies
Cerium-144	Ce-144	9469
Cesium-137	Cs-137	16.63
Iodine-131	I-131	8260
Krypton-85	Kr-85	2.114
Molybdenum-99	Mo-99	37061
Strontium-89	Sr-89	2670
Strontium-90	Sr-90	16.22
Xenon-133	Xe-133	21754
Yttrium-90	Y-90	0.31

Target production figures given in Table 2 are based on per gram – five day irradiation, so can be adjusted by target size and time of exposure. In operation, the system would be expected to operate on a 5 day on - 2 day off cycle. At the end of each 5 day operating period the Mo-99, Iodine and gaseous fission products would be extracted, fuel chemistry adjusted as needed, and then returned to the system for operation. Targets would be removed for shipment according to length of irradiation optimum for the specific isotope.

Table 2: Target Products: 5 day operation

Isotope	**Symbol**	**Curies/target gram**
Cadmium-109	Cd-109	0.086
Cobalt-60	Co-60	0.200
Dysprosium-166	Dy-166	0.002
Gold-198	Au-198	35.6
Gold-199	Au-199	0.50
Holmium-166	Ho-166	69.8
Iodine-125	I-125	72.8
Lutetium-177	Lu-177	59.2
Palladium-103	Pd-103	5.8
Rhenium-186	Re-186	33.0
Samarium-153	Sa-153	96.8
Selenium-75	Se-75	1.66
Tellurium-123m	Te-123m	0.195
Tin-117m	Sn-117m	0.50

CONCLUSION

The pumped fuel concept is the result of considerable design effort and operational experience gained over many years. The unique properties of liquid fuels combined with the transport of these fuels through a reactor core, rather than remaining in a static configuration, produce superior performance. The system is an intense thermal neutron source ideally suited to isotope production through fission product generation in the fuel and target irradiation. It represents a relatively low cost, low risk technology to produce large quantities of important radioisotopes.

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