Efficient and Timely Production Of Valuable Radioisotopes (in a Molten-Salt Reactor)

A. DeVolpi, PhD
Nuclear Applications Company
1371 Corte Paguera, Oceanside, CA 92057

ABSTRACT

Abundant and valuable Mo-99, tritium, He-3, and Pu-238 radioisotopes could be provided by a small multipurpose liquid-fueled molten-salt reactor (MSR). This enterprising means would satisfy high-priority national production and nonproliferation goals consistent with global threat reduction. It would conform to Congressional legislation requiring domestic, affordable, and proliferation-resistant radioisotope supplies for medical use, as well as meet increasing requirements having national-security applications. A single 10-100 MW(th) reactor, based on proven American technology, would fulfil growing deficiencies — at full cost-recovery, more likely a profit. Optimized for enhanced radioisotope production, the reactor would likely have to be government prioritized and located on a government reservation, in part to conform with global threat-reduction goals. It offers potential commercial product value of $billions/year, while reducing worldwide incentive and conditions for nuclear proliferation.

Contemporary Requirements
Ample and timely supplies of special radioisotopes — readily fulfilling objectives of the American Medical Isotopes Production Act — could be profitably produced in a single, small nuclear reactor.

Nuclear Applications Company, a California/Delaware small business, is promoting consideration of a small-scale dedicated molten-salt reactor (MSR) to create abundant quantities of crucial radioisotopes at competitive cost.

The proposal is based on decades of U.S. reactor development — some personally realized or witnessed at Argonne National Laboratory — for production of valuable radioisotopes, such as tritium (T-3), helium-3 (He-3), molybdenum-99 (Mo-99), and non-fissile plutonium-238 (Pu-238).

Because national-security-important isotopes would be generated, and in order to expedite licensing and construction, our patent application favors a privately-constructed, contractor-operated facility located on a government reservation.

This advantageous concept fits well into DOE/NNSA/NASA missions. The needed valuable radioisotopes, especially those in limited short- and long-term supply for medical and national-security applications, could be created and separated most efficiently, sufficiently, and economically in one molten-salt-cooled, low-enriched fluidized-fuel reactor.

By taking the initiative to develop U.S. domestic capacity for production, risk and incentive for international nuclear proliferation would be greatly diminished. This would be a proactive means of global-threat reduction, offsetting inevitable proliferation concerns that might arise in connection with growing shortages of special radioisotopes.

Legislative Foundation

The American Medical Isotopes Act of 2012 provides a statutory mandate to develop Mo-99 domestic supply that is secure, affordable, and reliable. It requires evaluation of a technology-neutral program, and encourages U.S. production of significant quantities for medical uses without using highly enriched uranium.

The proposed MSR is especially consistent with the 2005 Energy Policy Act, aimed at secure, affordable, and reliable sources of medical isotopes — particularly U.S. production of significant quantities of Mo-99 for medical uses — implemented in cooperation with non-federal entities.

The relevant legislation encourages commercial-scale projects for addressing potential civilian-use supply issues. Cooperation with non-federal entities is mandated for sharing costs in development, demonstration, and commercial application. A technology demonstration is favored for domestic production while also enhancing national security. DOE is to retain responsibility for final disposition of spent nuclear fuel and radioactive waste.

Our proposal conforms to global threat-reduction specifications for fuels, targets, and processes to supply domestic Mo-99 without using HEU. The U.S. government would retain responsibility for disposition of the much-reduced quantity of spent nuclear and radioactive waste created by irradiation, processing, and purification in an MSR. And the federal government would be constructively supporting development of medical-isotope generation that conforms to mandated non-proliferation goals.

Present Unfulfilled Requirements

Various international and national commissions, as well as Congress and NGOs, have recognized looming shortfalls in some essential radioisotopes. In the United States there are no major suppliers of Mo-99 for medical use, with nearly all of our domestic supply being furnished by foreign facilities.

Moreover, much of the world's supply of Mo-99 is derived mostly from very inefficient processes, including irradiation in research and test reactors that use highly enriched uranium (HEU) targets, which could provide materials for potential nuclear proliferation.

Production of almost all diagnostic and therapeutic radioisotopes in nuclear reactors is derived mostly from the fission process — either in reactor fuel or in specifically designed fissile targets. However, production is decreasing because of aging reactors, and because of the mandated shift to low-enriched uranium (LEU) with the need for full-cost recovery. Moreover, government subsidies are not available for all multipurpose isotopes, such as the fission product I-131.

Meanwhile, the production of tritium and helium-3 for U.S. national-security purposes has become increasingly expensive. In addition, there is a growing requirement for Pu-238 to satisfy long-term domestic national-security and international scientific-research missions.

Tritium. Nuclear weapons still require periodic replenishment, because tritium decays with 12.3-year half-life. Tritium reservoirs are emptied and precious depleted gas salvaged when the weapons are replenished or removed from service. Now that Cold-War tensions have relaxed, some nuclear weapons are being retired; therefore, previous production capacities are no longer anticipated. However, non-military controlled-fusion R&D is calling for increased quantities of tritium.

Helium-3. Its supply being dependent on recovery from nuclear weapons, the decay product of tritium is the rare gas He-3. It is especially useful in neutron detectors for domestic nuclear security throughout the world. It's also important for basic research applications. Some work-arounds minimize, but don't eliminate the usefulness of He-3.

Pu-238. The non-fissile radioisotope Pu-238, produced in high-power reactors from Np-237, is ideal for thermoelectric generators, especially for long NASA outer-space and NOAA deep-ocean missions.

Tritium, He-3, and Pu-238 all require government oversight and processing, partly in order to manage global-threat implications to U.S. national security. Because their supplies are becoming insufficient and expensive, prudent national-security and non-proliferation considerations advise U.S. government siting for a multi-purpose production facility with adequate capacity.

MSR for Radioisotope Production

To supply such specialized radioisotopes in a timely, cost-effective, and secure manner, a small molten-salt-cooled reactor is conceptualized: 100 MWth would be sufficient, using a single-fluid fuel/coolant (see conceptual diagram, Figure 1).

Some technical details about the MSR are provided later in this paper.

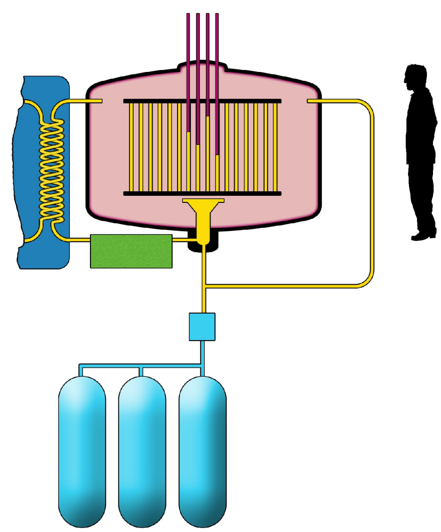

Figure 1. Small modular Molten Salt Reactor of this concept for production of valuable radioisotopes. Note large underground molten-salt emergency-dump tanks (rendered in light blue).

Compared to other reactors and to accelerators, an MSR is demonstrably the most efficient means of production: Fuel preparation is minimal; no solid-fuel or target fabrication is required; and nearly 100% duty cycle is achievable for irradiation and maximum efficiency in product extraction.

Alternative technologies are much less efficient and much more expensive per gram of isotope produced. Accelerators and accelerator-driven sub-critical reactors have inherent foil or sample irradiation target-density limitations; nevertheless, they have an indispensable and complementary role in producing other rare isotopes.

The MSR would yield two-to-three orders of magnitude higher radioisotope yield, with a smaller fissile loading, minimal cost, rapid production time, high efficiency, and versatile production. For each curie of Mo-99 generated in a liquid-fueled reactor, there is less uranium waste (by a factor of about 100) compared to yields from foil irradiation or solid-fuel reactors. All processes related to fabrication, irradiation, disassembly, and dissolution of solid-target foils are eliminated; therefore, radioactive waste management for the MSR is straightforward and less expensive, with comparatively low capital outlays and operating costs.

A domestically-built MSR would offer several other important benefits: high capacity and timely availability (five years or so, if given government priority and siting). It could have comparatively low construction cost, reduce government funding for national-security isotopes, and yield net income for the government and the facility operator, while not itself contributing to proliferation concerns.

Methods that do not involve fluidized-fuel reactors are necessarily much-less efficient and much-more costly per unit of radioisotope produced, even taking into account amortized cost. Liquid-fueled reactor systems would minimize or eventually eliminate the need for proliferation-susceptible, highly-enriched fissile targets.

Serious consideration has been given to two types of previously-developed fluidized-fuel reactors, one with fuel dissolved in aqueous solution, the other in molten salt (as in this proposal). Although the effective fission-product-production rate for aqueous-solution reactors might be equivalent to that achievable from an MSR, an aqueous system doesn't produce much tritium (and He-3) because lithium is not a major constituent. Also, aqueous-solution systems have serious limitations in safety and operational parameters.

Product yields of radioisotopes in solid-fuel reactors are limited by the means for which fission products can be extracted in a timely manner.

Irradiation of uranium foils or fuel in nuclear-reactors, while the predominant means currently in use, is highly inefficient because of interim decay during removal and processing cycles.

Accelerator generation is another order-of-magnitude less efficient because of the comparatively weak flux of neutrons. Accelerator-driven sub-critical reactors, lacking continuous processing of circulated solutions, would still have low yield. Accelerator-driven solution reactors might be better, but never as productive as the MSR.

All alternatives, however, potentially have useful and convenient roles for some specialized rare radioisotope creation.

Summary of MSR Production Expectations

From a broad U.S. economic perspective, one of the main advantages of a liquid-fueled radioisotope-production reactor is that product value would greatly exceed full-cost recovery.

Assuming operation at 100MWth, a single-fluid homogenous circulation MSR at a conservative 70% capacity factor should provide (with necessarily large product-yield and market-value uncertainties):

- Tritium production for U.S. government: ~210g/yr → **~$8.4M/yr**
- He-3 yield: ~100g/yr after 6 years of tritium decay → **(added value difficult to estimate)**
- Mo-99 6-day-Curie fission-product-yield: ~50g/yr → **~$140M/yr**
- Power at market value: ~100MWth → **~$12M/yr**

As a result, this small reactor would meet a significant portion of current U.S. requirements, reduce the federal-budget outlay for tritium, have an effective cost significantly less than other production proposals, and be self-supporting — even profitable.

Reducing Current Subsidies and Federal Outlays

The most prominent application for radioisotopes is in the medical field, for which production of almost all diagnostic and therapeutic radioisotopes comes from solid-fuel nuclear reactors, mostly as byproducts of the fission process. International and domestic industries, originally subsidized, now recognize the need for self-sustaining full-cost recovery.

The national-security radioisotopes tritium, He-3, and Pu-238 are currently funded by U.S. government national-security programs, and their production, processing, and disposition are carried out largely using government-controlled facilities.

MSR Technology Foundation

Two MSRs were built and operated decades ago successfully at Oak Ridge National Laboratory. They were licensed and operated by the Atomic Energy Commission, but were not designed as commercial isotope-production facilities. Oak Ridge originated liquid-fueled reactor research, and the two molten-salt reactors tested successfully .

The first, in the 1950s, was the Aircraft Reactor Experiment, 2.5 MW(th) military unit designed to attain high power density for use as an engine in a nuclear-powered bomber.

The second, in the 1960s, was the Molten-Salt Breeder Reactor Experiment (Figure 2), which operated intermittently for the equivalent of about 1.5 years full-power over a 4-year time frame. It was a 7.4 MWth test with an inherently safe neuronic "kernel" that simulated an epithermal liquid-fluoride thorium-breeder system.

The MSBR used single-fluid, non-breeding, low-power, molten-salt coolant with heat rejection to the air via a secondary (fuel-free) liquid salt. Piping, core, vessel, and structural components were made from highly durable and compatible Hastelloy-N alloy. The moderator was pyrolytic graphite, and the fuel solution was LiF-BeF2-ZrF4-UF4 (65-29-5-1). Secondary coolant was "FLiBe" (2LiF-BeF2). The reactor operated at up to 650 °C.

Experiments primarily involved two fissile-enriched loadings: first, uranium-235; later, uranium-233. (The U-233 was bred from thorium inventory of other reactors.) A breeder blanket of thorium salt was omitted from the MSBR experiments.

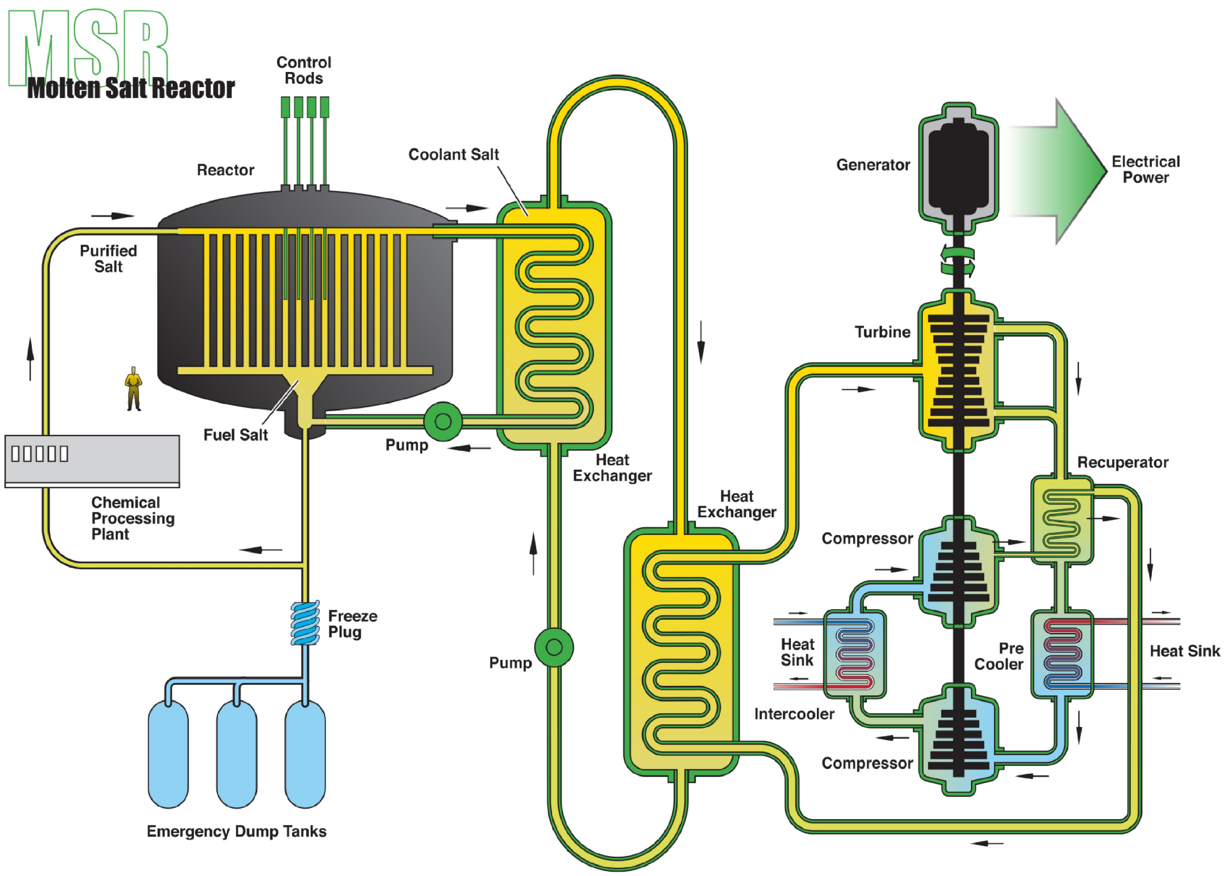

Figure 2. Diagram of molten-salt reactor based on 1960's experiment at Oak Ridge.

Reactor Fuel for Radioisotope Production

The proposed 100 MWth fluidized reactor for radioisotope production would require a relatively small inventory of uranium, just about 50 kg at 20% nominal enrichment (LEU). In a liquid-fueled system, suspended materials are extracted according to design and operating requirements, or they are continuously recycled through the core region to obtain more efficient burnup.

In contrast, in a typical solid-fuel reactor used for radioisotope production, only about 3 percent of the U-235 of a target foil is typically consumed. The remaining U-235, along with the other fission products and target materials, must be treated and disposed as nuclear waste.

Issues with Regard to Licensing and Siting

Reactor licensing has sometimes been a financial and procedural show-stopper, especially in the United States, and especially for non-traditional concepts.

If located on a government reservation, a privately-constructed and contractor-operated facility could benefit from expedited licensing and construction. That might be necessary in order to ensure timely compliance with Congressional mandates, and reduced limitations and expenses for national-security radioisotopes. Government siting is also justified in order to meet humanitarian priorities for needed radioisotopes.

Commercial contractors could manufacture and operate the reactor, supported by income from commercial-grade radioisotope sales in an open market. The greatly reduced costs resulting from government siting and licensing would also result in substantially lower federal budget outlays for the national-security isotopes.

Expedited licensing would be feasible because of (1) current national-security shortfalls in production of tritium, (2) homeland-security considerations in production of He-3, (3) government requirements for a long-term indigenous supply of Pu-238, and (4) long-standing precedents that have resulted in over 200 U.S. naval reactors having been built on a government reservation.

No U.S. operating license applications have ever been submitted that involve fluidized-reactor facilities in isotope production. Nor have nuclear regulatory bodies developed rules that could facilitate solution reactors for commercial isotope generation.

Aside from the two original AEC-licensed fluidized reactors, other liquid-salt or solution-type reactors throughout the world have been built, put into operation, or are being constructed subject to prevailing license requirements.

Specifics of MSR Radioisotope Production

A nuclear reactor nominally consumes about 1g of fissile material to generate about 1 MW of heat. Of that 1g consumed, about 6% would result in Mo-99 fission product. Fluid-fuel reactors are efficient and conducive to extraction of that important radioisotope, as well as other valuable byproducts. No other practical means comes close in efficiency of production and extraction .

In addition to the Mo-99, other radioisotopes used by the medical community can be processed efficiently: These include Sr-89, Xe-133, I-131, and Y-90. Higher specific activity of these isotopes in the MSR off-gas stream makes their retrieval much more effective compared to traditional uranium target-irradiation technology. For example, recovery of Sr-89 is about 1000 times more efficient compared to solid-target -irradiation of Sr-88.

When a lithium-based coolant is circulated through the reactor, tritium (and thus He-3) is simultaneously produced, at a rate which depends on reactor-design and lithium-enrichment parameters.

Important MSR Design Features

The MSR of this concept provide very high and efficient fissile conversion/burn up. Its compact core minimizes shielding, requires a comparatively small fuel requirement that results in minimal excess-fuel inventory.

Some unique, specific safety features are (1) the molten salt expands as temperature increases; (2) reactivity control is intrinsic to design passive safety; (3) a passive dump valve is made from a solid meltable-salt plug; (4) there are no energetic coolant reactions with H_2O; and (5) operation is at atmospheric pressure and minimal temperature.

In terms of nuclear-resources, the reactor could consume natural uranium or thorium to sustain its fuel cycle. The concept is amenable to the alternative fuel cycles, and it is adaptable to reutilization and transformation of fissile-depleted fuel.

Comparative Efficiency
Inherent in the now-prevailing target-irradiation method are inefficiencies such that only a small part (~0.4%) of available reactor uranium is used for Mo-99 creation. This leads to a high cost for Mo-99 supply and to an unnecessarily large quantity of slightly irradiated nuclear-fuel waste.

Since the fissile component of a fluidized reactor serves also as the homogeneous "target" for irradiation and generation of sought-after fission products, no separate fabricated target material is necessary. While the dissolved fuel may require periodic additions of LEU, it's anticipated that an original loading will be useable potentially for several decades. In comparison, uranium targets as fabricated for irradiation in a traditional reactor are expensive consumables.

Current practice requires the transport of an irradiated uranium target from the reactor to a processing facility for dissolution and materials recovery. This operation typically involves (1) an intermediate cooling step, (2) transfer equipment, and (3) an acid or base process to dissolve the target for product recovery. Such foil-processing steps involve personnel and equipment not needed for a liquid-salt fluid-fuel reactor system, wherein extracted isotopes are to be piped into a continuously operating isotope-separation device. This shortcut provides significant savings in cost of radioisotope extraction.

Special Governmental Considerations
DOE/NNSA is currently paying a high cost for tritium production in solid-fuel reactors, and is supply-limited in He-3 salvage from decayed tritium. In the long-term, increased national demand is expected for tritium, He-3, and Pu-238 — in addition to the statutory incentives for relieving Mo-99 domestic-supply problems.

Recognizing the value of advanced R&D on innovative small-reactor concepts, the DOE/NE web site has explicitly made reference to "liquid-salt" coolants that offer "added functionality and afford ability." In addition, DOE is providing grants that support prototype small modular reactors.

Nonproliferation actions have long been directed at limiting and controlling the quantity of HEU required for Mo-99 production. Removing U.S. dependency on foreign radioisotope sources of supply requires that several means be supported in order to obtain short-term and long-term resolution of medical-radioisotope shortfalls, as well as to consider limitations and expenses associated with national-security radioisotopes.

By taking advantage of American business initiative, intellectual-property rights, and nuclear technology, it's possible to further diminish incentives and justifications for other nations to develop proliferation-prone nuclear-production capability.

Development Work
In order to facilitate construction of the radioisotope-production reactor, several notable government and/or commercial development/demonstration issues would have to be addressed: radioisotope extraction efficiency and safety, material corrosion, leak minimization, and occupational-radiation management.

Optimized isotope production and separation would have to be established such that they assuredly provide public-service and national-security value, as well as meet or exceed current national requirements, preferably on a full cost-recovery basis. Importantly, U.S. development would pre-empt justifications for other nations to develop proliferation-prone isotope-production reactors.

Currently, there has been resurgence of international attention to liquid-salt and solution-type reactors, some with a thorium cycle, and some in the form of small prototype reactors. Several nations, particularly China, are now building MSRs, although not specifically for isotope production. Other national programs are in France, Norway, Czech Republic, India, Japan, and Russia. International GEN-IV attention includes concepts for power generation.

Several commercial MSR ventures have reached or progressed beyond conceptual design stages, including Transatomic, which emphasizes a 500 MW plant, fueled largely by light-water reactor waste. Flibe Energy is focusing on small modular system, Terrestrial Energy (a Canadian company) on heating tar sands, and ThorCon on an intermediate-power concept.

Summary

The proposed liquid-fueled fission reactor would be an enterprising approach that should satisfy several near-term high-priority national and commercial goals for supplying valuable radioisotopes. One small molten-salt reactor is sufficient to meet domestic requirements for essential medical and national-security radioisotopes.

Supported by congressional legislation, U.S. government agencies have overtly recognized and acted upon the need to develop domestic supplies of at least one valuable medical radioisotope, Mo-99. Despite past circumstances wherein options have been limited for new reactors, the proposed concept offers near-term radioisotope-production benefits while avoiding most shortcomings.

This is based on the molten-salt reactor originally developed at Oak Ridge. If optimized for enhanced radioisotope production, the new version would circulate liquid-salt compounds that provide very good and relevant operational properties, functioning at low (near-atmospheric) pressure. These features result in reduced mechanical stress, simplified construction, safe operation, and significantly reduced radioactive waste. The fluidized reactor of this proposal likely would have to be government prioritized and best located on a government reservation.

The small reactor of this concept would appear to produce timely and sufficient radioisotopes, meeting or exceeding current national requirements — at the very least on a full cost-recovery basis, more likely at a profit. It offers a potential commercial product market value of many billions of dollars per year, while reducing incentive and justification for global nuclear proliferation.