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PRODUCTION AND COLLECTION OF HE-3 AND OTHER VALUABLE ISOTOPES USING Mu*STAR

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Abstract

We propose an example facility based on Mu*STAR, an accelerator-driven molten-salt-fueled graphite-moderated thermal-spectrum reactor that can operate with different fissile fuels and uses a LiF-BeF2 molten eutectic carrier salt. In the first example, we propose using the Li-6 in the LiF carrier to produce more than 2 kg/y of tritium (decaying to He-3 with 12.3 year half-life) using a 2.5 MWb superconducting proton linac to drive the subcritical 500 MWt reactor burning surplus plutonium. The collection of other valuable fission-product radioisotopes like 133Xe will also benefit from the high temperature and continuous removal and separation afforded by fractional distillation.

INTRODUCTION

There are many uses for He-3 that are related to fundamental scientific sudies, but one use that requires large quantities is for instrumentation for the US Department of Homeland Security to detect plutonium in cargo at the many entry points into the US. Because of the inadequate supply of He-3, the DHS has had to use less optimum neutron absorber material such as boron-10. The project proposed here is to develop a new method to produce large quantities of tritium, even more than is needed to maintain the US nuclear weapons stockpile, in order to guarantee a continuous and copious supply of He-3.

A MU*STAR ADSR SOLUTION

The resurgence of molten-salt-fueled nuclear reactor designs, after almost 50 years since the pioneering Molten-Salt Reactor Experiment (MSRE) at ORNL, has inspired this new method to produce and collect radioisotopes and He-3. Compared to traditional light water reactors, new molten-salt designs operate at higher temperature and allow access to fission products by means of helium flow over or though the core of an operating reactor without the constraint of cladded solid fuel. In the method described here, the helium picks up the volatile isotopes and carries them to a hot cell facility where fractional distillation and chemical techniques are used to separate, collect, and store each isotope, thereby purifying the helium gas before it is recirulated though the reactor. Thus, the core of the reactor will contain almost a factor of a million fewer volatile radioactive isotopes than other designs and may not require a containment vessel.

We describe below an example facility based on Mu*STAR, an accelerator-driven molten-salt-fueled graphite-moderated thermal-spectrum reactor that can operate with different fissile fuels and uses a LiF-BeF_2 molten eutectic carrier salt. Preliminary simulations indicate that with the correct isotopic fraction of ^{6}Li in the LiF carrier salt, the pilot plant reactor will produce about 2.5 kg/y of tritium (decaying to ^{3}He with 12.3 year half-life) using a 2.5 MW_{beam} superconducting proton linac to drive the subcritical 500 MW_t reactor burning surplus plutonium.

The high operating temperature of the molten reactor in the Mu*STAR sytem, and the contiuous removal of the tritium from the reactor, imply low partial pressure to minimize escape and embrittlement issues. The collection of other valuable fission-product radioisotopes like Xe-133 will also benefit from the high temperature and continuous removal and separation afforded by fractional distillation. In this project, in addition to the study of ^{3}He production, we will also investigate possibilities to collect radioactive isotopes that are naturally produced as fission products as well as those that can be generated by suitable additions to the molten-salt fuel. We will estmate production rates as well as probabilities of collection considering factors that are dependent on the chemistry of the MS fuel, fission products, and reactor and distillation construction materials. We outline the components of a pre-conceptual design of this system.

ISOTOPE PRODUCTION ISSUES

Helium-3:

He-3 that is used in the US is primarily from the decay of tritium that is stored for the nuclear weapons stockpile.

$$^3_1T \rightarrow ^3_2He + e^- + \bar{\nu}_e$$

The tritium needed for the nuclear stockpile is now provided by a combination of recovery from decommissioned weapons and from new production at the Watts Bar TVA reactor in Tennessee. The method used is to replace fuel rods in the reactor with Tritium Producing Burnable Absorber Rods (TPBARs) that contain Li-6. The neutrons in the reactor core then convert the Li-6 to T.

$$n + {}^6_3Li \rightarrow {}^4_2He\ (2.05\ MeV) + {}^3_1T\ (2.7\ MeV)$$

The TPBARs remain in the reactor for an 18 month cycle until they are removed during a regular reactor refueling and sent to SRNL where they are opened to recover the tritium. The tritium is put into metal hydride storage beds [1] where the He-3 “weeps” out and is recovered.

Current US industrial consumption of helium-3 is approximately 60,000 liters (approximately 8 kg) per year (in 2014); cost at auction has typically been approximately $100/liter although increasing demand has raised

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prices to as much as $2,000/liter in recent years [2]. Thus At $100/liter 8 kg He-3 is $6M, or 750,000/kg or $750/g. At $2,000/liter, it is $15,000/g. So at the highest price, the 8 kg/y of He-3 would be worth $120,000,000 per year.

By comparison, it is estimated that the US government pays about $300,000/g for tritium in large part because US laws require that components for the nuclear stockpile be domestically sourced and controlled. At that price, 2.8 kg/y would bring $840,000,000 per year, which is 7 times more than the 8 kg/y He-3 would bring.

One important fact is that the collection of the tritium and He-3 together could amount to almost a billion dollars of annual revenue. A second is that the tritium is worth 7 times the He-3.

Tritium:

The status of the US Nuclear Weapons Stockpile is recently described in the NNSA's 2018 Nuclear Stockpile Stewardship and Management Plan (NSSM) [3]. In it the arguments and some of the costs for upgrading the methods for producing tritium needed for the US stockpile are found:

- The Y-12 Li-6 enrichment facility needed for TPBARs is obsolete
  - Cost unknown to update.
- At least one additional reactor is needed to produce the total of 2.8 kg/y of T needed after 2025
  - At that time the weapon decommissioning will end, along with that source of recycled T
  - Additional reactors need to be upgraded and certified for TPBAR use
- A new domestic source of enriched uranium fuel is needed to supply the reactors
  - Sources of weapons components must be domestically located and controlled
  - A uranium enrichment plant is envisioned for a cost of >$2B

In addition, the two Watts Bar reactors (and other potential ones at Sequoyah) are not on NNSA sites, but are located at a commercial location under the purview of the Nuclear Regulatory Commission (NRC). The NRC has already imposed limitations on the number of TPBARs that can be loaded into the Watts Bar reactor because of unacceptable tritium losses into the environment. This uncertainty implied by having a national nuclear security function operating within a commercial reactor is also a problem, but is not discussed in the NSSM.

The opportunity for the community is that the plan described below addresses each of these needs at reduced cost and increased security and provides a path for extra T production for even more He-3. By building a relatively small pilot plant on the SRS controlled by the NNSA/DOE and Environmental Management (EM/DOE) we can use onsite surplus plutonium as fuel. One simulation indicates that about 230 kg of plutonium will produce 2.8 kg of T using the natural abundance Li-6/Li-7 ratio in the LF component of the eutectic molten salt fuel.

A cost estimate made at ANL for a 25 MWb (beam power), 1 GeV CW proton accelerator using all required DOE factors for contingency etc, done in 2015 came to $800M. Using magnetron power sources that are being developed by Muons, Inc. instead of klystrons or IOTs that were in the estimate would decrease that cost by $170M [4]. To scale that design down by a factor of 10 in beam power to the 2.5 MWb needed for the simulation described above is an exercise that is yet to be done. Simulations indicate that 650 MV will work almost as well as 1 GeV. So we are pretty confident that the accelerator for a pilot plant will cost around $400M.

Using the same materials as were used in the MSRE, the 500 MWt Mu*STAR reactor is estimated to cost $300M to cover component costs for graphite, fuel, pumps, and container.

Additional costs for the hot cell recovery of the volatile elements from the reactor purge gas are yet to be estimated, but it we believe that the cost to produce tritium that is envisioned in this project is less than the $2B needed just for the uranium enrichment facility described in the NSSM.

Other Fission Products

We will investigate the possible recovery of other radioisotopes such as Xe-133 now used for diagnosis and treatment of lung cancer. We believe there are many such isotopes that may make the method described here unusually productive for life-saving and industrially-interesting applications.

MU*STAR ISOTOPE COLLECTION

Mu*STAR is a graphite-moderated, thermal-spectrum, molten-salt-fueled reactor that is operated using an external accelerator to direct protons onto an internal spallation target. GEM*STAR can be operated with many fuels, without redesign, for process heat and/or for electricity generation. Figure 1 shows its basic components, where the active volume is 93% graphite (gray) and 7% molten salt (red) made up of an appropriate eutectic mixture of lithium, uranium, plutonium, and/or thorium fluorides with a melting point above 500 C. Safety features of the design include the 500 MW_{th} power output design limit, corresponding to not needing 1) a critical mass of fissile material for operation and 2) "defense in depth" measures for loss-of-coolant accidents since the heat generated by decays of fission products after the accelerator is turned off can be dissipated by passive external air cooling.

Helium flow over the hot core removes volatile isotopes and carries them to a hot cell where they are separated out chemically or cryogenically with the fractional distillation column indicated in Figure 1 and then safely stored while they decay. This reduces the inventory of volatile isotopes in the reactor by a factor of almost a million compared to reactors used at Fukushima.

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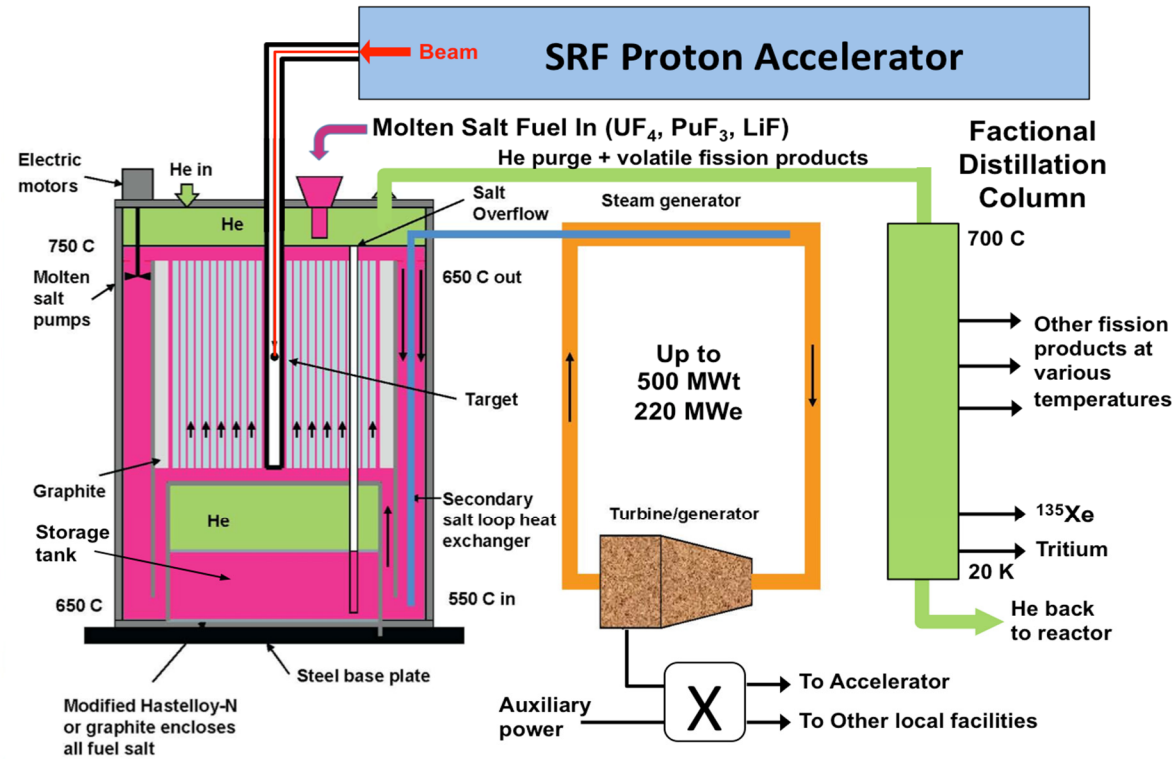

Figure 1: Conceptual view of a Mu*STAR accelerator-driven molten-salt-fueled subcritical graphite-moderated nuclear reactor configured to produce electricity and valuable radioisotopes. The beam energy and power shown in the figure correspond to burning PuF_3 in a eutectic LiF and BeF_2 carrier salt. One of the features of the Mu*STAR design is that volatile radioactive isotopes are continuously removed from the reactor by passing a flow of helium through it. The tritium that is collected in a hot cell, shown as a fractional distillation column in the figure, will be stored in an underground chamber at the reactor site where the He-3 will be collected. We expect that users of tritium at the NNSA, Fusion Reactors, DHS and other places will also collect the He-3.

FUTURE WORK

Up to now, molten-salt reactor simulators have used enriched Li-7 because they regarded tritium production from the Li-6 component of the LF euectic to be undesirable as tritium releases from reactors have led to restrictions by regulating bodies like the NRC. We, on the other hand, are interested in producing and collecting as much tritium as possible and so our technical objectives in Phase I proposal to the DOE SBIR program will focus on the techniques that will let us do that safely, without uncontrolled escape, and effectively such that the He-3 can be recovered from the stored tritium. Working downstream from the accelerator to the recovery of the He-3, the problems that we intend to address are listed below.

1. Producing Tritium:
   a) Accelerator vacuum to reactor helium purge gas interface Low beta focus at the interface with plasma lens and differential pumping
   b) Beam rastering on internal spallation target depleted uranium cooled by the 700 C MS Fuel [5]
   c) Optimise tritium production, investigate use of natural abundance of lithium isotopes in LF.
   d) MuSim [6] simulations of Mu*STAR burning surplus plutonium
2. Collecting tritium:
   a) Containment in the reactor without embrittlement or escape low partial pressure, correct materials & chemistry
   b) Out of core and into the purge gas by controlling the salt chemistry
   c) Into the hot cell to be isolated and stored
   By fractional distillation or chemical means
3. Collecting He-3
   a) Underground storage facility for all isotopes
   b) Metallic Hydride Beds for tritium storage

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