HANDLING GEM*STAR VOLATILE RADIOACTIVE FISSION PRODUCTS

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Abstract

One of the promising future reactor designs with accelerator-produced neutrons is GEM*STAR, which is a subcritical thermal-spectrum reactor operating with molten salt fuel in a graphite matrix. GEM*STAR is able to use natural uranium as well as spent fuel from light water reactors (LWR). The capture of volatile fission products is required for the system. The volatiles caught in the helium gas circulating around the reactor core can be trapped in bottles. Numerical simulations have been performed to estimate the amount of fission products that will need to be confined when spent nuclear fuel from LWR is used.

INTRODUCTION

Accelerator Driven Systems (ADS) have been proposed for several missions in advanced nuclear fuel cycles since the 90's, where a source of neutrons produced by proton-induced nuclear fragmentation or spallation is delivered to the nuclear core. Minor actinide destruction through transmutation is one mission of ADS, and significant reduction of minor actinides (MA) is expected. One of the promising future reactor designs with accelerator-produced neutrons is GEM*STAR (Green Energy Multiplier*Subcritical Technology for Alternative Reactors), developed by Accelerator Driven Neutron Application (ADNA), which is a subcritical thermal-spectrum reactor operating with molten salt fuel in a graphite matrix. This reactor is able to use natural uranium as well as unreprocessed spent fuel from light-water reactors (LWR), generating as much electricity as the LWR had generated from the same fuel. The advanced design of GEM*STAR using liquid nuclear fuel is quite different from LWR that use solid nuclear fuel that stores fission products inside of its fuel rods. At each step in the GEM*STAR fuel cycle, there is no need for any chemical separations of the fuel.

Fission Products - Health Concern

Commercial LWR reactors use ceramic fuel pellets that are enclosed by a zirconium alloy sheath that capture the radioactive isotopes produced by fission reactions. These fuel rods accumulate volatile radioactive fission products during years of operation. On the other hand, liquid molten-salt fuel requires a novel confinement system for fission products in GEM*STAR. The fuel salt is confined by modified-Hastelloy-N and graphite. On the surface of the fuel salt, helium gas is continuously flowing to remove noble gases and other volatiles. The helium gas circulates around the reactor and carries the volatiles out of the reactor where centrifuges or cryogenic methods can remove the radioactive volatile elements from the helium. The removed volatiles can be safely stored in bottles distant from the reactor, where they can decay over time. The design of the volatile radioactive element removal system is essential to ensure the safety of molten-salt reactors because many of such elements are also health concerns, as shown in Table 1.

Table 1: List of Health Concerns

Nucleus	Half-life	GI absorption
^{131}I	8.0 days	100%
^{137}Cs	30 yrs	100%
^{90}Sr	28 yrs	30%
^{140}Ba	13 days	5%

This paper presents our estimates for volatile fission production from the burn-up of LWR spent nuclear fuel by spallation neutrons from heavy metal targets generated by energetic proton beams [1].

SIMULATION OF THE REACTOR

Nuclear reactions in GEM*STAR with LWR spent fuel were simulated with MCNP5 code to obtain k_{eff} for a neutron flux of 9×10^{21} n/cm^2 in the reactor core, and ORIGEN2 code for evolution of the isotopic distribution. The details of GEM*STAR parameters are presented in Ref [2]. The outside dimension of the core is a cube of 503 cm on a side with an internal 50-cm thick graphite reflector on all six sides. GEM*STAR will have a molten-salt intake flow of about 0.8 l/h of UF_4. The calculation accounts for only a simple isotopic evolution of a single load of fuel. As starting nuclides, a spent nuclear fuel mixture was taken for a burn-up of 3.3 GWd/t of uranium dioxide that had been out of the LWR for 10 years [3].

Burning of Actinides

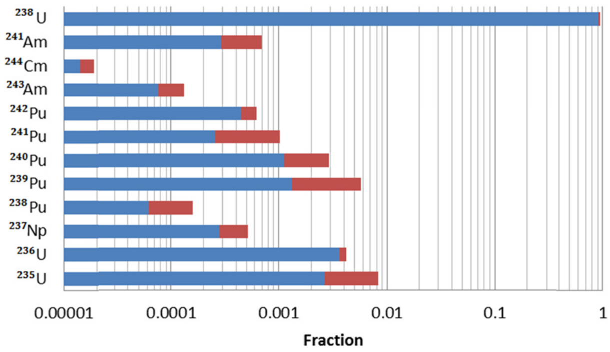

Figure 1: GEM*STAR burn-up of LWR spent fuel.

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The isotopic fractions of the actinides in the fuel are presented for the calculation at $k_{eff} = 0.99$ in Figure 1. The fractions are normalized so that the sum of the actinides in used LWR fuel equals 1. The whole bar (blue+red) is the initial fraction in the fuel, while the red part is the fraction participating in neutron-induced reactions or nuclear decays in GEM*STAR.

It should be noted that the red bar is not the direct reduction factor of actinides because the minor actinides are not only reduced but also produced in the reaction chains. However, the fissile isotopes of ^{235}U and ^{239}Pu are clearly reduced by nuclear fission with the thermal neutron capture. This result indicates the benefits of recycling the used LWR fuel in GEM*STAR, where the amount of fissile uranium and plutonium not burned in the LWR is reduced without chemical reprocessing.

Isotopic Distribution of Fission Fragments

The isotopic distribution of fission fragments is shown in Figure 2. The fractions are normalized such that the sum of the initial actinides equals 10^{12}. The sum of fission fragments and final actinides is more than 10^{12} because nuclear fission splits the fissile isotope into two nuclei.

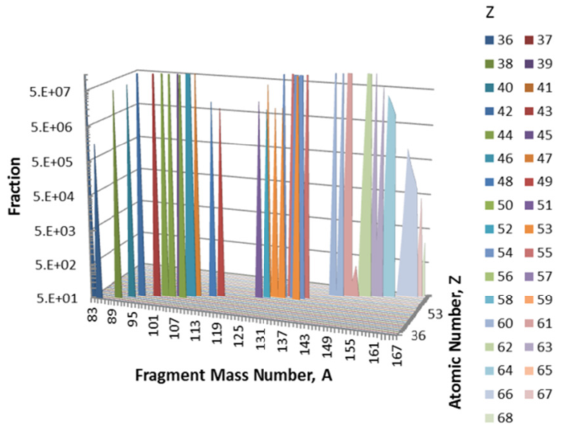

Figure 2: Isotope distribution of fission fragments from burn-up of LWR spent nuclear fuel in GEM*STAR.

Fission product yield by mass for thermal neutron fission of LWR spent nuclear fuel is shown in Figure 3, as the projection of the isotope distribution. The double peak structure of mass yield originates from the fact that the major fission products are produced from both ^{235}U and ^{239}Pu.

The yield is dependent on the parent atom and the energy of the initiating neutron. In this calculation, one peak occurs at ^{99}Tc while the other peak is at ^{134}Xe.

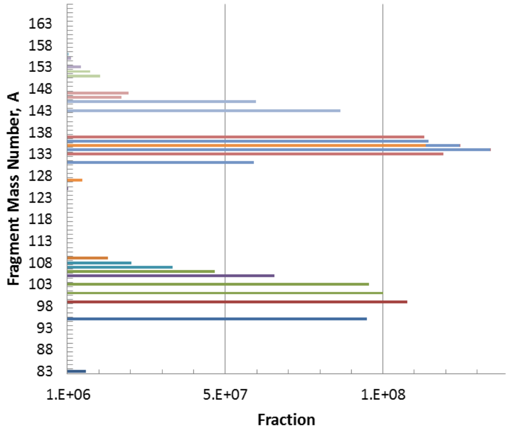

Figure 3: Mass yield of the fission products, obtained as the projection of isotope distribution.

Volatile Fission Products

The amount of the volatile radioactive fission products is estimated in Table 2. The weight of each isotope is calculated based on grams per metric-ton of used LWR fuel.

Table 2: Volatile Fission Products

Nucleus	Half-life	Weight [g]	Activity [Ci]
^{85}Kr	10.8 yrs	5.97E-01	2.33E+02
^{90}Sr	28.9 yrs	2.01E+01	2.768E+03
^{103}Ru	39 days	4.24E+01	4.90E+03
^{106}Ru	374 days	2.15E+01	3.27E+06
^{125}Sb	2.75 yrs	6.99E-01	7.27E+02
^{127}Te	9.35 hrs	5.37E-01	1.62E+02
^{131}I	8.02 days	9.59E+00	1.19E+06
^{135}I	6.57 yrs	6.59E+01	2.66E+04
^{134}Cs	2.07 yrs	3.25E-03	4.184E+00
^{135}Cs	2.3 M yrs	6.30E-02	7.252E-05
^{137}Cs	30.2 yrs	6.66E+01	5.757E+03
^{135}Xe	9.2 hrs	6.18E+00	6.498E+05

The structure of GEM*STAR (500 MWt or 220 MWe) and the molten-salt technology suggest the amount of total inventories in the reactor is equivalent to 76 metric tons as uranium mass. The total weight of volatiles listed in Table 2 is about 230 g, corresponding to 17 kg of volatile fission products in GEM*STAR. The storage of this amount of volatiles is easily achieved in storage bottles.

The volatile fission products presented in Table 2 were selected from such isotope list for traditional LWR. There could be more volatiles from the molten-salt fuel because the operating temperature of fuel salt in GEM*STAR is about 700 °C, being significantly higher than that of

LWR. The circulating helium gas also carries volatile stable fission products. The amount of volatiles captured in storage bottles is probably higher than estimated in Table 2.

Future of ADS and Accelerator Technology

The energy output is mainly based on the nuclear fission of ^{235}U and ^{239}Pu. The latter is supplied not only from the initial actinides but also from ^{238}U (actually 73% of initial amount) with fission neutrons and additional neutrons from accelerators. The principal advantage of ADS in comparison with critical reactors is greater flexibility with respect to fuel composition. The present calculation of GEM*STAR that uses unreprocessed spent fuel from LWR is a good example to demonstrate the great performance of ADS, including one of its missions to produce fissile materials for subsequent use by irradiating fertile elements [4]. The flexibility of fuel composition depends on the amount of supplemental neutrons from accelerators. Deeper burning of spent nuclear fuel is enabled if the proton beam intensity is increased. Thus, accelerator technology is the key to expand the possibility of ADS technology.

SUMMARY AND CONCLUSIONS

The ADS reactor using molten-salt fuel is a very promising future technology for burning many kinds of nuclear materials including natural uranium, excess plutonium from weapons, very abundant thorium, and used LWR fuel, with safety advantages of sub-criticality. Volatile fission products are continuously purged by a circulating helium gas. The volatile radioactive element removal system is essential to ensure the safety of molten-salt reactors because many of such volatile elements are also health concerns. The amount of volatiles was estimated from the present simulation in GEM*STAR and they are able to be stored in the storage bottles with reasonable costs.

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