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Development of a Nondestructive Assay Design Concept for Molten Salt Reactor Material Accountancy

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ABSTRACT

Molten salt reactors (MSRs) are a popular design concept for Generation IV nuclear reactors. Some variations in MSR design concepts that have been proposed concern the fuel cycle, liquid or solid fuel, coolant salt constituents, etc. One popular MSR design approach utilizes a liquid $^{232}Th/^{233}U$ fuel cycle. The design characteristics of liquid-fueled MSRs present many unique safeguards challenges. These include the presence of new diversion pathways due to in-line fuel processing (removal of fission products, online refueling, ^{233}Pa separation/decay), difficulties in accounting for changes in fuel composition at different locations in the fuel pathways, and complications involving in-line nondestructive assay measurements taking place in transient, corrosive, and high-neutron background environments. The objective of this design concept is the development of an in-line safeguards system for liquid-fueled MSRs by identifying possible diversion pathways and utilizing effective nondestructive assay techniques to accurately assess the movement of special nuclear material in a facility. This was accomplished using state of the art codes in MSR burnup depletion modeling to model operational parameters and radionuclide inventory of a representative liquid $^{232}Th/^{233}U$ fuel reactor. The fission product composition of the fuel following burnup has been analyzed to identify potential isotopes for nondestructive assay analysis. The gamma spectra of the resulting fission products were identified. Following the identification of the isotopes of monitoring interest, a thermal and neutronic analysis was conducted to determine the required setup for an effective detection system in the MSR environment. Various materials, detectors, and configurations will be proposed and detection of the characteristic gammas from the fuel's fission products will be evaluated for utility in nuclear material accountancy monitoring. Accurate knowledge of the location and amount of these elements is an essential part of preventing the proliferation of nuclear material towards undeclared military or malicious security activities.

INTRODUCTION

In 1957, the International Atomic Energy Agency (IAEA) was established as an independent body to promote peaceful uses and prevent military use of nuclear materials. The stated technical objective for the IAEA international safeguards program is "the timely detection of diversion of significant quantities (SQ) of nuclear material from peaceful nuclear activities to the manufacture of nuclear weapons or of other nuclear explosive devices or for purposes unknown, and deterrence of such diversion by the risk of early detection" [1]. SQ of nuclear materials are those that could potentially be used to produce a nuclear weapon. The SQ of various isotopes of nuclear materials are given in Table 1 [2].

Table 1. Significant quantities of nuclear material as defined by the IAEA.

Material	Quantity
Direct Use Material	
Plutonium	8 kg Pu
Uranium-233	8 kg ^{233}U
Highly Enriched Uranium	25 kg ^{235}U
Indirect Use Material	
Uranium	75 kg ^{235}U
Thorium	20 MT Th

In order to achieve the safeguards technical objectives set forth by the IAEA, detection systems must be put into place that can account for all nuclear material present in a nuclear facility. While Comprehensive Safeguards Agreements (CSA) have been established with the IAEA concerning inspection and detection measures for material accountancy at the facilities in operation today, nuclear reactors of the future may offer additional challenges for detection or pathways for diversion of nuclear materials.

A popular design concept for Generation IV nuclear reactors is the molten salt reactor (MSR). The defining feature of MSRs is the use of molten salt as the primary reactor coolant or fuel mixture. Molten salts are desired due to their high heat capacity, low vapor pressure, and other desirable neutronics properties. Most of the proposed MSR designs operate without pressurization at temperatures from about 500 °C to about 1400 °C (773.15 K to 1673.15 K). This is in marked contrast to a pressurized water reactor (PWR), which operates at about 315 °C (588.15 K) under a pressure of 15.2 MPa [3]. Operating at high temperatures without pressurization allows for a greater thermal efficiency and eliminates a possible source of radiation dispersal due to pressure transients. A variety of MSR designs have been proposed with variations in the neutron spectrum they operate in, the loop configuration, and the state of fuel used. One design concept is the proposal of a liquid ^{232}Th/^{233}U fuel cycle thermal MSR in which new ^{233}U fuel is bred from neutron capture in the ^{232}Th.

A liquid ^{232}Th/^{233}U fuel cycle is vastly different from a traditional fixed and solid fuel cycle, making current safeguards techniques incompatible for accurately monitoring nuclear materials. Safeguards challenges that result from a liquid fuel cycle include unique challenges in accounting for changes in fuel composition at different locations in the fuel pathways, the presence of new diversion pathways due to in-line fuel processing (removal of fission products, online refueling, ^{233}Pa separation/decay, etc.), and intricacies associated with real-time, in-line nondestructive assay (NDA) taking place in transient and corrosive environments.
While MSRs are widely recognized as a favorable design for next generation nuclear reactors, the safeguards systems currently in place are not yet prepared to verify declarations of nuclear materials from these facilities. Theoretical work surrounding different MSR designs and the necessary information for producing an effective safeguards-by-design system for various MSRs is a major focus of research around the world today. The development of a theoretical framework for new safeguards policies and material accountancy methods in MSR environments will be an essential factor in eventually making the MSR a reality [4,5].

APPROACH

A preliminary model MSR was created and modeled in SCALE. The initial model assumed that the fuel had reached an equilibrium concentration and was operating at a steady state. A safeguards system based on IAEA timeliness and material quantity goals was developed taking into consideration the environment in which it must operate. This environment will be determined by the parameters of the MSR model. The design of the safeguards system was modified as necessary to achieve the material accountancy goals. After the safeguards system was modeled in SCALE, the generated source term was implemented in MCNP to produce a detector response model and subsequent gamma spectrum. This gamma spectrum was used to identify candidate gammas with distinct energies to be used in identification of ^{233}U fission.

MSR Model

Each of the assumed parameters in Table 2 were specified for the simulated MSR in order to identify effective NDA techniques, possible diversion pathways, and material accountancy changes [6].

Table 2. Design parameters for model MSR

Neutron Spectrum	Thermal neutron spectrum
Driver Material (Initial Fissile Isotope)	^{233}U as main driver material
Configuration	Single-Fluid, Two-Region
Separation of Fissile Material	Online separation of fissile material
Refueling	Online Refueling
Protactinium Separation	Online separation of Pa from the salt
Fuel Salt Base	^{7}LiF, BeF_2, ThF_4, UF_4

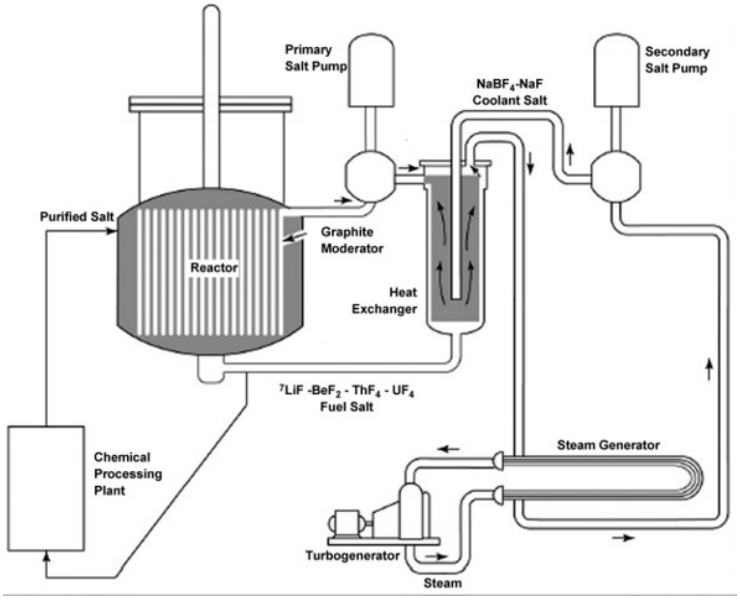

Figure 1. Reference MSBR design used for modeling [7]

Figure 1 shows the single-fluid, two-region Molten Salt Breeder Reactor (MSBR) that was used for the model in designing the safeguards system. A thermal neutron spectrum (graphite moderator) was chosen because thermal neutrons are ideal for breeding ^{233}U from ^{232}Th. Driver material is used to start the neutron chain reaction in the reactor core. The initial fuel loading in a MSBR requires the presence of enough fissile material to power the reactor until the ^{233}U concentration reaches equilibrium. The driver material type affects diversion pathways and possible undeclared material that could be produced because it could introduce additional fissile materials, such as ^{235}U or ^{239}Pu, to the facility [8]. These isotopes have their own verification signatures and safeguards requirements. The driver material also determines what safeguards measures might be necessary to achieve safeguards technical objectives. ^{233}U is the ideal fissile material to start up a thorium-based MSR because the salt composition with this initial fissile material is closest to the equilibrium fuel composition. The reference MSR is a single-loop model that will be developed with a single-fluid molten salt mixture consisting of ^{7}LiF, BeF_2, ThF_4, and UF_4. The model will account for separation of ^{233}U from the fuel salt. This ^{233}U will then be used to refuel the reactor in an online refueling process.

Placement of Detection System

The detection system was placed inline after the fuel had passed through the core and through the heat exchanger but before chemical reprocessing. At this point of the reactor the temperature of the salt is the coolest while the salt still has the greatest concentration of fission products before they are extracted from the mixture. The detection system being placed near this pipe will produce a gamma spectrum from the characteristic photons emitted by the fission products of ^{233}U.

Materials

Hastelloy N is the surface contact material for the nearly all metal surfaces in contact with the molten salt material for this design. Hastelloy N was specifically designed to withstand use in high neutron-temperature corrosive environments [7].

Thermal Shielding

Since the detector was placed in the reactor cell it was necessary that a system was designed to maintain the detector at an adequate operating temperature in a high temperature environment. The proposed design to keep the detector within operating temperature range included a stainless steel (SS316) structural enclosure with a ceramic insulation lining and an active cooling refrigerant (Figure 2).

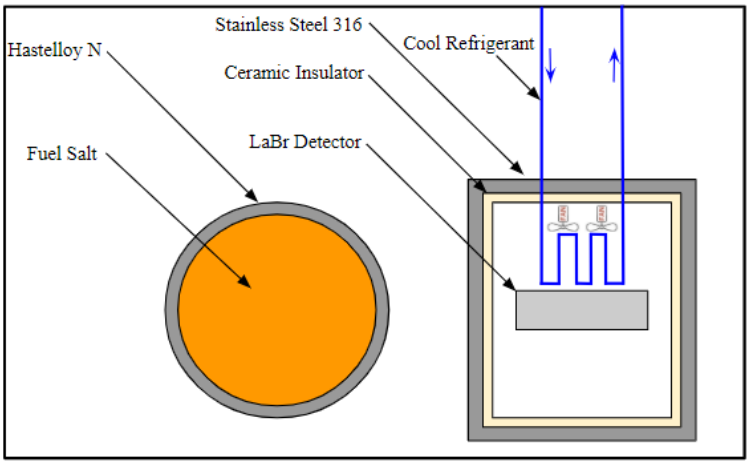

Figure 2. Visual representation of the cooling system with detector shielding and location

The assumptions made for the thermal analysis were that the outside of the SS316 shielding for the detector had a surface temperature of 538 °C (810.93 K). Also, the inner surface temperature of the ceramic insulation was evaluated at an assumed 38 °C (310.93 K). With this assumption, the detector would not reach the max operating temperature. The resulting calculation predicted the minimum amount of heat removal required for the detector system to operate was approximately 1000 W. A refrigerant cooling system capable of withdrawing 3000 W of heat was chosen to ensure that the detector was properly cooled.

SCALE Computations

The source term for modeling the gamma spectrum from the MSR fuel salt was produced using SCALE software from ORNL. Transport Rigor Implemented with Time-dependent Operation for Neutron depletion (TRITON) and Oak Ridge Isotope Generation and Depletion (ORIGEN) codes within SCALE were used to generate cross section files and gamma and neutron spectra respectively. The generated cross section file was used as an input for the ORIGEN depletion calculations.

The fuel in TRITON was set up as a 2-dimensional view of the pipe with one quarter of the pipe's cross section being defined. The boundary in the fuel was defined to have an isotropic return for the particles. The fuel was defined with chemical compounds for ^{7}LiF, BeF_2, UF_4, and ThF_4. The fuel occupies a radius of 19.05 cm with Hastelloy N piping that is 2.54 cm thick. Depletion calculations were run for the fuel salt mixture at 566 °C (839 K) for the time of 12 months. TRITON compiled a file for the cross sections of all the elements based on the defined environment.

ORIGEN was used to run point depletion calculations. The initial depletion calculations were run at a thermal power of 30 MWth for 12 months of operation. Initial values for the material quantities of the fuel salt (Table 3) rates were defined, and feed and removal rates were provided for the given power by an example SCALE input [9]. ^{232}Th was being fed into the system with various fission products being removed. ORIGEN was then used to produce spectra for emitted gammas and neutrons at various time intervals.

Table 3. Fuel salt composition by component [7]

Component	Atomic Fraction	Weight Fraction	Grams
^{7}Li	0.28351	0.07852	1.335E+06
Be	0.06327	0.02251	3.826E+05
F	0.60459	0.45341	7.708E+06
Th	0.04745	0.43462	7.389E+06
^{233}U	0.00071	0.00655	1.113E+05
^{235}U	0.00012	0.00110	1.871E+04
^{234}U	0.00027	0.00252	4.284E+04
^{236}U	0.00008	0.00077	1.315E+04

Two additional source term generation cases were simulated in ORIGEN for comparison with the initial deletion case. In the first variation, the initial quantity of ^{233}U by mass in the fuel salt was doubled from the initial case. In the second variation, the initial quantity of ^{233}U remained doubled, and the thermal power of the simulation was increased to 325 MWth. These cases were referred to as the Typical Case, the Doubled U Case, and the Increased Power Case, respectively.

MCNP Simulations

After development of the prospective source terms using SCALE, the gamma energies and their respective intensities as predicted by the ORIGEN point depletion models were used to create source spectra in MCNP. The ORIGEN output data provided values for gamma energies and intensities at multiple time steps during the depletion simulations. In this analysis, the gamma information was retrieved for cases after three months and twelve months of depletion. This depletion time was selected to accommodate the IAEA timeliness detection goals for the diversion of one SQ of irradiated direct use material such as ^{233}U.

The initial setup of the MCNP model consisted of a lanthanum bromide ($LaBr_3$) detector placed perpendicular to a cylindrical source meant to represent the molten thorium fuel salt in a Hastelloy N pipe (Figure 3a). Source particles were generated in the simulated fuel salt and biased to travel in the direction of the detector to reduce computation time (Figure 3b). A thermal shield was added to the $LaBr_3$ input model to account for any potential impact on the detector response due to attenuation in the thermal shield (Figure 3c).

For each source term produced by ORIGEN, an MCNP input was created using the respective gamma energies and probabilities. An F8 energy deposition tally was used on the cell representing the $LaBr_3$ detector crystal to tally the photons that would be detected by the setup.

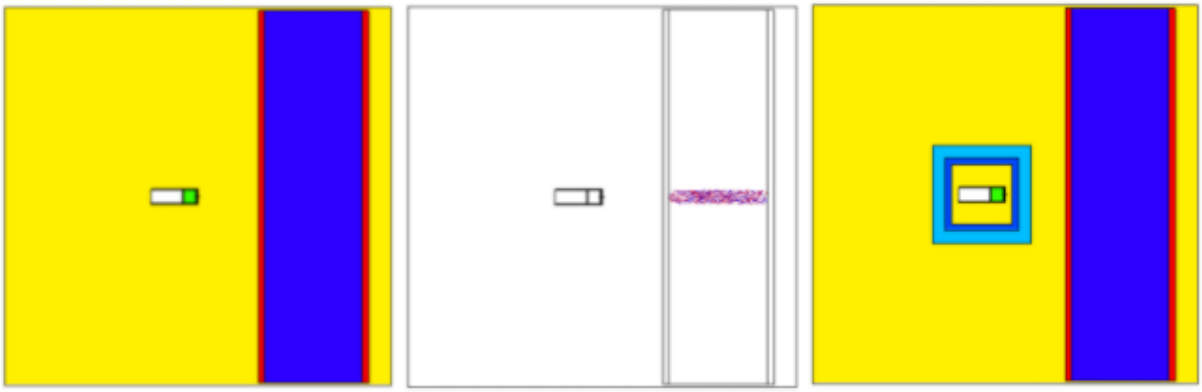

Figure 3 (a). Initial XY plot of system geometry with detector and fuel in Hastelloy N pipe. (b). Source points shown in color. Particles were biased to travel in the -X direction. (c). System geometry after addition of thermal shield.

RESULTS

Each of the F8 tallies produced in MCNP was split into 1024 energy channels spaced at linear intervals between 0 and 3.5 MeV. For each of the cases modeled, the photon tallies produced by MCNP reported relative uncertainties of less than 5% for each energy channel below 2 MeV. The relative intensities of energy deposition in the detector were binned in each channel and used to produce a gamma spectrum for each case. An unshielded photon tally was generated to identify candidate gamma peaks for detecting irradiated ^{233}U fuel (Table 4). The energies corresponding to the best resolved peaks were identified and compared with characteristic gammas of various radioisotopes to identify the potential fission products responsible for the gamma signatures observed.

Table 4. Characteristic gammas identified in MCNP detector response models

Isotope	Gamma Energy (keV)
^{125}Sb	603
^{130}I	743
^{131m}Te	849
^{91}Sr	1030
^{166}Ho	1380
^{138}Cs	1430
^{38}Cl	1600
^{9}Be	1750
^{90}Y	2180

After identifying potential gamma energies of interest, the detected intensities of each gamma source term were retrieved from the MCNP outputs. These intensities were then compared (Table 5) to determine if the different cases resulted in significant and detectable changes in the output gamma spectra.

Table 5. Comparisons of average gamma intensity per keV for each depletion case

Three Month Depletion Cases			
Case	**Comparison to Typical Case**	**Comparison to Doubled U Case**	**Comparison to Increased Power Case**
Typical Case	-	-7.60%	-6.84%
Doubled U	+9.12%	-	+1.06%
Increased Power	+8.12%	-0.72%	-
12 Month Depletion Cases			
Case	**Comparison to Typical Case**	**Comparison to Doubled U Case**	**Comparison to Increased Power Case**
Typical Case	-	-8.18%	-0.79%
Doubled U	+9.69%	-	+8.61%
Increased Power	+1.19%	-7.28%	-

CONCLUSIONS

After only three months of depletion, variations in the parameters of the initial fuel composition and the operating characteristics of the model MSBR produced observable differences in the resulting gamma spectra. Applying this design concept to additional cases with varying parameters and modeling source terms from extended depletion calculations could amplify the observed differences. The average differences in gamma intensity per keV between the typical operation case and the two modified cases were greater than the relative uncertainty reported by MCNP. The two modified cases, which both featured the same initial ^{233}U content, did not demonstrate significant differences in gamma intensity per keV after three months of depletion. After 12 months of depletion, the differences between the average gamma intensities of the typical and doubled U spectra were statistically the same as the differences observed after three months of depletion. However, the variations between the typical and increased power cases were not statistically significant after 12 months. This demonstrates that variations in the operating parameters impact the resulting gamma spectra from the fission fragments and could be used to conceal improper material processes such as excess initial quantities of ^{233}U.

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