Antineutrino Monitoring of Thorium Reactors

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Various groups have demonstrated that antineutrino monitoring can be successful in assessing the plutonium content in water-cooled nuclear reactors for nonproliferation applications. New reactor designs and concepts incorporate nontraditional fuels types and chemistry. Understanding how these properties affect the antineutrino emission from a reactor can extend the applicability of antineutrino monitoring. Thorium molten salt reactors (MSR) breed 233U, that if diverted constitute a direct use material as defined by the International Atomic Energy Agency (IAEA). The antineutrino spectrum from the fission of 233U has been estimated for the first time, and the feasibility of detecting the diversion of 8 kg of 233U, within a 30 day timeliness goal has been evaluated. The antineutrino emission from a thorium reactor operating under normal conditions is compared to a diversion scenario by evaluating the daily antineutrino count rate and the energy spectrum of the detected antineutrinos at a 25 meter standoff. It was found that the diversion of a significant quantity of 233U could not be detected within the current IAEA timeliness detection goal using either tests. A rate-time based analysis exceeded the timeliness goal by 23 days, while a spectral based analysis exceeds this goal by 31 days.

I. INTRODUCTION

The accident that occurred at the Fukushima-Daiichi nuclear power plant following the 2011 earthquake and subsequent tsunami in Japan led to an international effort to increase the development of accident tolerant nuclear fuels specifically to withstand a core meltdown in the case of a beyond-design-basis event such as a loss of coolant. This has led to a revival in the interest in thorium molten salt reactor (MSR) designs. Despite the improvement to reactor safety, the production and online reprocessing of 233Pa which decays into 233U creates a state-sponsored proliferation risk. Current IAEA methods used to detect the divergence of nuclear material are relatively intrusive and cannot directly measure the fissile inventory of the reactor 1. Moreover, the safeguards approach for this new class of reactors has not yet been defined. Groups at Lawrence Livermore National Laboratory (LLNL) and Sandia National Laboratories (SNL) have shown that using antineutrino detectors provide a remote and non-intrusive method to observe the diversion of nuclear material 2. In this paper the feasibility of using antineutrino monitoring to detect the diversion of 233U from a thorium MSR is evaluated.

II. BACKGROUND

Antineutrinos are weakly interacting neutral particles produced from the beta decay of neutron rich fission products. Unlike neutrons, beta particles, and gamma rays: antineutrinos can be detected from well outside the reactor core because of their low interaction cross section. About six antineutrinos are produced from each fission resulting in an antineutrino flux of about 1023νe/s from a 1 GW reactor. Antineutrino monitoring of traditional nuclear reactors uses the varying spectral contributions of fission products from 235U and 239Pu over time to make the detection of materials diversion feasible. When these isotopes fission they produce different beta emitters with various yields. The antineutrino spectrum for each fissile isotope is an combination of the antineutrino spectra of individual fission products, with each contribution fixed by the fission yield and branching ratio. As a result, each fissile isotope has a distinct antineutrino spectra shape and amplitude making its growth or depletion potentially accessible through antineutrino monitoring. For thorium reactors the difference in antineutrino spectra from 233U and 235U, the most prominent fissile isotopes, allows for diversion detection.

A. Antineutrino Detection

Antineutrinos can be detected through their interactions with quasi-free protons in a liquid scintillating medium, using the inverse beta decay process shown in Eq. 1.

\[
\nu_e + p \rightarrow e^+ + n \quad Q = -1.804 \text{ MeV}
\] (1)

The threshold for this reaction is 1.804 MeV, limiting detection of antineutrinos to those above that energy. In a liquid scintillator detector, the positron created from this reaction will slow in the detector and annihilate with an electron. The positron will deposit almost all of its energy at the end of its trajectory, and because subsequent annihilation occurs instantaneously, the energy of
the positron and the gamma rays emitted from annihilation create an indistinguishable prompt signal. In a gadolinium-doped scintillator, the neutron captures on the Gd dopant within a few tens of microseconds, depending on the dopant concentration, a delayed signal from the resulting 8 MeV gamma ray cascade is formed. These two signals, detected in close time coincidence defines an antineutrino event.

In the present work, the antineutrino emission from the reactor is studied using a design similar to that of a detector developed and tested at LLNL [3]. The detector is assumed deployed at a 25 meter standoff, 10 meters below the surface, equivalent to a water overburden of about 25 meters. At these depths a previous iteration of this detector, SONGS1, experienced a singles and delayed coincidence background count rate of 3725 and 105 counts a day respectively for a target mass of 0.64 tons [2]. For a 3.6 ton detector the uncorrelated daily background rate is assumed to be 21,000 counts. This uncorrelated background may be subtracted, thereby contributing a 150 count uncertainty on the signal. Due to significant improvements in background rejection from the muon veto system, larger fiducial volume, and inter-event timing cuts the correlated background is found to contribute an additional 200 counts per day [3]. It is assumed these counts are evenly distributed within 500 keV energy bins. While the backgrounds cannot be precisely known without an actual deployment, these estimates represent a reasonable extrapolation based on the measured properties of the 3.6 ton detector, and the known signal and background for the SONGS detector. Other experiments have shown similar signal to background ratios [3].

Although the antineutrino flux from the reactor is about $(10^{21} \text{ν}/\text{s})$, the low interaction cross section for this reaction ($\sigma = 9.52 E^2 \times 10^{-44} \text{cm}^2$) significantly reduces the number of events detected [5]. Figure 1 gives a visual representation of the interaction cross section and antineutrino spectra effect on antineutrino detection for $^{235}$U and $^{233}$U fissions, as well as the ratio between the two. The falling antineutrino spectrum coupled with the quadratic increase in the interaction cross section results in a unique detection spectrum for each fissionable isotope [6]. The sum of their antineutrino spectra can be used to estimate the inventory of fissionable content in a given reactor throughout the cycle.

### B. Thorium Reactors

A thorium MSR core consists of a seed and blanket separated by a graphite moderator. Neutrons emitted from fissile material contained in the seed, converts $^{232}$Th in the blanket to $^{233}$U. At the beginning of the reactor lifetime the seed is comprised of low enriched uranium (20% $^{235}$U); over time enough $^{233}$U is bred through the blanket to be used in the seed. $^{233}$U is formed from neutron capture by $^{232}$Th and subsequent beta decay chains shown below:

$$^{232}\text{Th}+n \rightarrow ^{233}\text{Th} \rightarrow ^{233}\text{Pa} \rightarrow ^{233}\text{U}$$  \hspace{1cm} (2)$$

Online reprocessing occurs in the blanket to separate $^{233}$Pa and $^{233}$U from $^{232}$Th. As shown in Figure 2, salt from the blanket undergoes flourination to re-inject uranium into the core, and minimize reactivity losses. $^{233}$Pa is then extracted from the salt, and allowed to decay in a sub-critical storage containment. Another reduction process is used to extract $^{232}$Th and replace it in the blanket. After $^{233}$Pa decays to $^{231}$U, it is reintroduced into the core [7]. Because the conversion ratio for $^{232}$Th

| Property         | Quantity    |
|------------------|-------------|
| Target Mass      | 3.6 Tons    |
| Efficiency       | 39.2 $\pm$ 4.7\% |
| Energy Resolution| $\sqrt{E/(\text{MeV})}$ |
| Fiducial Volume  | 100\%       |
| Standoff         | 25 Meters   |
| Overburden       | 25 M.W.E    |
| Signal to Background | 1262/200  |

### TABLE I. Summary of detector parameters used in this analysis.
and $^{233}$U is less than one for this reactor, enriched uranium is also added to the seed as makeup fuel. If a state diverts $^{233}$Pa before its daughter product can be reintroduced into the core, enough $^{233}$U could be proliferated to construct a nuclear device.

The reactor used in this analysis is a 500 MWth thermal thorium MSR taken from the Sandia National Laboratories Nuclear Fuel Cycle Catalog [8]. The fuel evolution was calculated using ORIGEN 2.2, a reactor neutronics software to simulate the isotopic inventory of nuclear systems [9]. The fissile inventory of the core over the reactor’s lifetime is shown in Figure 3. The inability to fully refuel the core and the material degradation from using molten salts reduces the lifetime of these reactors to a few decades.

FIG. 2. (Color online). Flow diagram of a thorium MSR reprocessing unit showing the movement of fissile and fertile material. The introduction of thorium and enriched uranium is also added to the seed as makeup fuel. If a state diverts $^{233}$Pa before its daughter product can be reintroduced into the core, enough $^{233}$U could be proliferated to construct a nuclear device.

III. ANTINEUTRINO EMISSION

To determine the expected antineutrino emission from a thorium MSR, the antineutrino spectra for $^{231,235,238}$U and $^{239,241}$Pu must be evaluated. Because of the graphite moderator the fast neutron flux in the blanket is negligible. As a result, fissile actinides such as $^{233}$Th and $^{233}$Pa do not contribute to the antineutrino emission from the reactor. The antineutrino spectra from $^{235,238}$U and $^{239,241}$Pu have been previously determined by converting the measured electron emission following neutron irradiation at ILL and FRM II [10, 11]; while the antineutrino spectra from the fission of $^{233}$U is not available in open literature, and must be estimated through an aggregate summation method.

The antineutrino spectrum from the fission of an isotope, $N(E_\nu)$, is a combination of spectra, $P_\nu(E_\nu, E_0, Z)$, from its fission products. In this work, the fission yield and uncertainties was taken from ENDF-349 [12].

$$N(E_\nu) = \sum_n Y_n(Z, A, t) \times \sum_i b_{n,i}(E_0) P_\nu(E_\nu, E_{0i}, Z)$$

(3)

Where $Y_n(Z, A, t)$ is the number of beta decays per second for a given isotope $(Z, A)$; $b_{n,i}(E_0)$ is the branching ratio to an excited state with the electron energy spectrum endpoint:

$$E_{0i} = Q_n - E_{ex}$$

(4)

Here $Q_n$ is the $Q$ value for the beta decay of isotope $(Z, A)$, and $E_{ex}$ is the excitation energy in the daughter nucleus $(Z + 1, A)$. The decay information for all the fission products was taken from the Evaluated Nuclear Structure Data Files [13]. The fission products for which the decay data was absent were neglected because they only represented less than $10^{-6}$ of the cumulative fission yield, or had such large $Q$ values that these nuclei would be beta delayed neutron candidates such that the antineutrinos emitted from their decay would be less than 3 MeV (below the energy range of this assessment).

The spectrum shape factor, $P_\nu(E_\nu, E_0, Z)$ shown in Equation 5 is derived from the phase space relationship between the electron and antineutrino and the normalization constant, $k$, for each branch: as well as the Fermi Coulomb function, $F(E_0, Z)$, which accounts for the Coulomb attraction between the emitted electron and daughter nucleus is shown in Equation 6 [14]. An allowed Gamow-Teller spectral shape was assumed for all beta decays.

$$P_\nu = k E_\nu^2 (E_0 - E_\nu)^2 F(E_0, Z)$$

(5)

Additional corrections to the antineutrino spectra include both the electromagnetic and weak-interaction finite size correction, the radiative corrections, and the weak magnetism correction [6, 10, 15, 16]. Effects of the screening correction, were neglected do its small contribution to the antineutrino shape [10]. Previous assessments show multiple methods of evaluating the spectrum.

FIG. 3. (Color online). Fission fraction of fissile isotopes throughout the reactor’s lifetime with associated uncertainties from simulation. At the beginning of the reactor’s lifetime, most of the fissions comes from $^{235}$U. Over time the contribution of $^{233}$U to the fission fraction increases until it reaches an equilibrium.
IV. DIVERSION SCENARIO

Once the antineutrino spectra of fissile isotopes were calculated, the corresponding detection rate, $D(E_{\bar{\nu}})$, was found using Equation (6). Here $\rho_p$ is the proton density, $V$ is the detection volume, $\epsilon$ is the detector efficiency, $T$ is the counting time, $r$ is the distance from the detector to the reactor core, and $\phi$ is the antineutrino rate derived from the fission rate in the reactor [2]. The survival probability, $P_{ee}$, which accounts for the oscillation of neutrino flavor is also accounted for. However, because the detector is located 25 m from the reactor core the loss of antineutrinos due to oscillations was negligible.

$$D(E_{\bar{\nu}}) = \frac{T \rho_p V \epsilon (E_{\bar{\nu}}) \phi(E_{\bar{\nu}}) P_{ee}(E_{\bar{\nu}}, r)}{4\pi r^2}$$  (6)

Once the antineutrino is emitted there is no way to determine which fissile isotope was responsible for its emission. Regardless, the count rate evolution and overall detected antineutrino energy spectrum will still reflect changes in inventory of fissile material in the reactor. The IAEA set limits of concern for unaccounted nuclear material that can be used for the construction of a nuclear device. In the case of $^{233}$U, the IAEA defines a significant quantity to be 8 kg with a timeliness detection goal of 30 days. Two cases are compared in the diversion study: a baseline scenario in which the reactor is under standard operation, and an anomalous scenario in which a state is trying to divert $^{233}$U. The thorium molten salt reactor used in this analysis reaches a $^{233}$Pa production.
TABLE II. Approximated $^{233}$U antineutrino spectra, and correlated uncertainties using the summation method.

| $E_\nu$ [MeV] | $N_\nu$ [$\nu\text{ fission}^{-1}\text{ MeV}^{-1}$] | $\sigma$ [%] |
|---------------|---------------------------------|----------|
| 2.00          | $9.45 \times 10^{-1}$          | 9.03     |
| 2.25          | $7.76 \times 10^{-1}$          | 10.0     |
| 2.50          | $6.34 \times 10^{-1}$          | 6.85     |
| 2.75          | $5.14 \times 10^{-1}$          | 9.39     |
| 3.00          | $4.10 \times 10^{-1}$          | 11.6     |
| 3.25          | $3.22 \times 10^{-1}$          | 13.3     |
| 3.50          | $2.54 \times 10^{-1}$          | 14.6     |
| 3.75          | $2.20 \times 10^{-1}$          | 13.2     |
| 4.00          | $1.58 \times 10^{-1}$          | 11.6     |
| 4.25          | $1.25 \times 10^{-1}$          | 10.6     |
| 4.50          | $9.90 \times 10^{-2}$          | 7.52     |
| 4.75          | $7.92 \times 10^{-2}$          | 12.5     |
| 5.00          | $6.25 \times 10^{-2}$          | 16.5     |
| 5.25          | $4.88 \times 10^{-2}$          | 18.7     |
| 5.50          | $3.77 \times 10^{-2}$          | 23.8     |
| 5.75          | $2.83 \times 10^{-2}$          | 23.6     |
| 6.00          | $2.07 \times 10^{-2}$          | 27.1     |
| 6.25          | $1.45 \times 10^{-2}$          | 36.7     |
| 6.50          | $9.62 \times 10^{-3}$          | 23.5     |
| 6.75          | $5.92 \times 10^{-3}$          | 19.9     |
| 7.00          | $3.53 \times 10^{-3}$          | 19.6     |
| 7.25          | $2.20 \times 10^{-3}$          | 24.5     |
| 7.50          | $1.36 \times 10^{-3}$          | 22.1     |
| 7.75          | $7.61 \times 10^{-4}$          | 17.6     |
| 8.00          | $3.91 \times 10^{-4}$          | 19.9     |

In this analysis the antineutrino count rate evolution in an anomalous scenario was compared to the expected
antineutrino counts under standard operations (baseline) using a hypothesis testing procedure [4]. Figure 5 shows the count rate evolution in both the baseline and anomalous scenarios along with the LS regression fit. The antineutrino count rate for this thorium MSR shows a smaller decline over time than that of the PWR seen in Reference [4]. This MSR is refueled online with more frequency than a PWR, as a result the reactivity of a reactor, for a given thermal output, is primarily controlled by the makeup fuel; whereas, in a PWR the boron concentration of the coolant throughout a cycle indiscriminately affects the actinides fission rate.

Once the antineutrino count rate was determined, a quadratic fit using least square (LS) regression was applied to the rate. To eliminate dependencies between the coefficients, the LS regression was performed on the sample mean \((\bar{t} - \bar{t})\) as shown in Equation 8.

\[
N_t^{(B)} = \beta_0^{(B)} + \beta_1^{(B)} (t - \bar{t}) + \beta_2^{(B)} (t - \bar{t})^2
\]

Here the superscript are consistent with those used in the spectral analysis. Comparing the coefficients \(\beta_0, \beta_1,\) and \(\beta_2\) between the measured and baseline fit will indicate if material has been diverted using the following hypothesis test:

\[
H_0^i : \beta_i^{(M)} = \beta_i^{(B)} \quad \text{versus} \quad H_a^i : \beta_i^{(M)} \neq \beta_i^{(B)}
\]

Equation 9 can be determined using the following test statistics,

\[
s_i = \frac{\hat{\beta}_i^{(M)} - \hat{\beta}_i^{(B)}}{\sqrt{\sigma^2(\hat{\beta}_i^{(M)}) + \sigma^2(\hat{\beta}_i^{(B)})}}
\]

and the corresponding \(\textit{p}\) value:

\[
p_i = 2 \cdot P(S \geq |s_i|)
\]

Here \(S\) has a Student’s \(t\) distribution with \(2 \cdot (n - 3)\) degrees of freedom, with \(n\) being the number of count rate measurements and \(\sigma\) referring to the uncertainties in the daily count rate. Although not included in this work, the systematic uncertainties in the detector response and antineutrino rate may be reduced if previous measurements on this reactor can allow for template matching [4].

When applying the test to the count rate evolution, two of the three coefficients reject the null hypothesis in favor of the alternative hypothesis. The one coefficient, \(\beta_0\), that showed no statistical difference was related to the absolute antineutrino rate emission. This shows that the test is not dependent on precise knowledge of the expected count rate, but observes general trends (i.e. is the count rate monotonically decreasing or not) in the rate evolution.

To determine the robustness of the hypothesis tests to statistical variations in LS coefficients, a Monte Carlo simulation was used to generate one hundred thousand anomalous and baseline detected antineutrino count rate evolutions assuming a Gaussian distribution. An ideal threshold for the true positive/false positive rate was identified as 95%/5% . A true positive result was identified as the test correctly identifying an anomalous scenario, while a false positive result indicated that the baseline was incorrectly identified as an anomalous scenario. Figure 7 shows the ROC curve for the rate analysis under a 55 day counting period as well as the counting time required to reach the desired TP/FP rate. Under the 55 day constraint, an 81%/5% TP/FP rate is achieved. The desired rate is achieved by increasing the counting time to 78 days.
**TABLE III. Sensitivity of the count rate evolution test using Monte Carlo simulations.** The results show the false positive rate that would incur for a 5% false positive rate for both the diversion scenario presented in the study, and a misreported power shift.

| Counting Time | Diversion Scenario TP/FP | Misreported Power TP/FP |
|---------------|--------------------------|-------------------------|
| 55 Days       | 81%/5%                   | 58%/5%                  |
| 78 Days       | 95%/5%                   | 73%/5%                  |
| 114 Days      | 99%/2%                   | 95%/5%                  |

When this procedure was applied to pressurized water reactors to observe the diversion of plutonium, it could not identify anomalous activity as quickly if the operator misreported the thermal power output [4]. Because of the transient growth of the antineutrino spectra caused by the diversion of $^{233}$U, misreporting the reactor power would not mask proliferation. Figure 6 shows the expected antineutrino count rate if the reactor was operating at a 7MW excess. Here the $\beta_0$ coefficients are matched, requiring more time to detect material diversion. After applying the same procedure, as mentioned above, an anomalous scenario can be detected within 114 days in the presence of a false power report. Additionally, the effects of misreported power shifts can give insight to the test’s sensitivity to detector drifts. The detector used in this analysis has not been deployed long enough to study these effects, but the SONGS1 detector showed a less than 1% drift while taking data [21].

**V. CONCLUSION**

The antineutrino emission from a thorium MSR was analyzed to determine if the diversion of a significant quantity of $^{233}$U could be observed within the IAEA timeliness goal. In order to perform the analysis, we calculated the antineutrino emission spectrum of $^{233}$U, based on the fission product yields. Using a spectral and rate-time based analysis the diversion $^{233}$U could be detected 61 and 53 days after the diversion of 8 kg of $^{233}$U respectively; in a total counting period of 86 and 78 days. Usually a spectral analysis is more sensitive to diversion than the rate-time analysis because it provides more information about the antineutrino emission. In this analysis the integral spectrum over the entire 55 day period is being binned into a single histogram so the day to day changes in the rate of each bin is averaged out.

Evaluating the antineutrino count rate gives insight to the power level of the reactor, where analyzing the detected antineutrino spectra is a reflection of its isotopic concentration. Although this method requires less time, relying solely on the count rate makes the analysis susceptible to operator malfeasance. When using the antineutrino count rate, there is a trade off between exceeding IAEA timeliness goals and being sensitive to false declarations of reactor power.

The test for material diversion was heavily dependent on counting statistics. Desirable test performances are attainable through feasible improvements to antineutrino detectors, such as better instrumentation for small scale antineutrino detectors or increasing the detector mass. Additionally, variations in thorium reactor designs may also affect the sensitivity to diversion. Some models incorporate scheduled refueling, reducing the counting time from 55 days to 30 days; or utilize epithermal and fast neutron energies, which would effect the antineutrino emission from fission. Other designs incorporating solid fuels do not separate the $^{233}$Pa from the blanket. This changes the SNM from unirradiated direct use material to irradiated use material, allowing for longer diversion detection constraints.

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