

**Antineutrino monitoring of burning mixed oxide plutonium fuels**

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**Background:** Antineutrino monitoring of reactors is an enhanced nuclear safeguard that is being explored by several international groups. A key question is whether such a scheme could be used to verify the destruction of plutonium loaded in a reactor as mixed oxide (MOX) fuel.

**Purpose:** To explore the effectiveness of antineutrino monitoring for the purposes of nuclear accountability and safeguarding of MOX plutonium, we examine the magnitude and temporal variation in the antineutrino signals expected for different loadings of MOX fuels.

**Methods:** Reactor burn simulations are carried out for four different MOX fuel loadings and the antineutrino signals as a function of fuel burnup are computed and compared.

**Results:** The antineutrino signals from reactor-grade and weapons-grade MOX are shown to be distinct from those from burning low enriched uranium, and this signal difference increases as the MOX plutonium fraction of the reactor core increases.

**Conclusion:** Antineutrino monitoring could be used to verify the destruction of plutonium in reactors, although verifying the grade of the plutonium being burned is found to be more challenging.

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**I. INTRODUCTION**

Mixed oxide (MOX) fuels refer to reactor fuel that contain oxides of more than one fissionable actinide, and typically involve a mixture of plutonium and natural or depleted uranium. When the plutonium makes up about 5% of the total MOX fuel, the fuel burns with a similar reactivity to that of normal low enriched uranium (LEU) fuels. One major motivation in burning MOX fuels is the destruction of either weapons-grade or reactor-grade plutonium. However, such schemes raise new nuclear safeguard concerns. For example, is it possible to verify how much MOX fuel was loaded, and is it possible to verify the grade of plutonium being burned? A number of current and past studies [1] have examined antineutrino monitoring of reactors to verify the reactor power and the isotopic content of the fuel. The antineutrinos are emitted in the beta decay of the fission fragments, and the number of beta decays per fission is dependent on which actinide is fissioning. Thus, in principle, the temporal variation in the number of antineutrinos emitted by the reactor core probes variations in the isotopic content of the reactor fuel as it burns. Here we examine the feasibility of using antineutrino monitoring to verify the burning of plutonium in MOX fuels.

MOX fuels have been used in thermal reactors since the 1960s and today many commercial thermal reactors are loaded with one-third MOX. Some advanced light water designs are capable of accepting 100% MOX loadings. In this work we examine the antineutrino signatures from burning MOX, varying the amount and the grade of plutonium being burned. The potential for antineutrinos to monitor the destruction of MOX fuel mainly lies in the fact that fissioning  $^{239}\text{Pu}$  only emits about 43% (66%) as many antineutrinos per fission as does  $^{238}\text{U}$  ( $^{235}\text{U}$ ) (Table I). Thus, the magnitude of the emitted antineutrino flux and its variation with the fuel burn-up could, in principle, verify that plutonium is the dominant actinide being burned, provided that the reactor power is known. Here we compare the antineutrino signals for MOX loadings of

33.3% and 100% of the total reactor core, either reactor-grade or weapons-grade plutonium. In all cases, the MOX is taken to be 5.3%  $\text{PuO}_2$  and 94.7%  $\text{U}_{\text{natural}}\text{O}_2$ . When the MOX fuel is 33.3% of the core, the remaining fuel is taken to be fresh 2.56% enriched  $\text{UO}_2$ .

**II. REACTOR BURN SIMULATIONS**

Our reactor simulations are carried out using the MonteBurns code [2], which couples the Monte Carlo neutron transport code MCNP [3] to the burn code CINDER'90 [4]; the latter uses 63 neutron energy groups and tracks up to 3400 nuclides, including 638 isomers. As a benchmark, we first consider a standard H. B. Robinson Unit 2 (HBR2) PWR fuel assembly that had 2.56% enriched fresh LEU and no MOX fuel. Samples from fuel assemblies have been assayed and the spent fuel isotopics measured in detail [5]. The sample chosen in the present work, assembly BO-5 sample N-9B-N, was irradiated to a burn-up of 23.81 GWd/MTU and the detailed power history recorded. The power history involved four burn cycles, with shutdowns of the order of 1–2 months between cycles. We reproduce the reported history using variable concentrations of burnable boron poison rods over the four burn periods. We then predict spent fuel isotopic inventories for the uranium and plutonium isotopes within about 5% of measurement.

There are many MOX loadings that could be considered, and in the present study we consider four specific loadings. In the first two, the entire reactor core is assumed to be MOX fuel, one reactor-grade (RG) and one weapons-grade (WG) plutonium. The initial isotopics are given in Table I. The second two cases that we study assume that these MOX fuels make up one-third of the core; the remaining two-thirds of the fuel is assumed to be fresh 2.56% enriched LEU, the same as in the HBR2 test case. In simulating the MOX fuels, the average power history of the HBR2 assembly is retained.

TABLE I. Initial isotopics for both MOX fuels. The last row lists the antineutrino detection cross section per fission for each actinide in units of  $10^{-43} \text{ cm}^2$  [6,7] [Eq. (1)].

Isotope (%)	$^{235}\text{U}$	$^{238}\text{U}$	$^{238}\text{Pu}$	$^{239}\text{Pu}$	$^{240}\text{Pu}$	$^{241}\text{Pu}$	$^{242}\text{Pu}$
Weapons	0.68	94.01	–	5.0	0.3	–	–
Reactor	0.68	94.01	0.203	2.653	1.373	0.535	0.535
$\int \sigma(E_{\bar{\nu}})S(E_{\bar{\nu}})dE_{\bar{\nu}}$	6.61	10.10	–	4.34	–	5.97	–

*Entire MOX cores.* Figure 1 displays the nuclide fission contributions during burn-up of pure RG and WG MOX fuels. As expected, the largest contribution to the fissions is  $^{239}\text{Pu}$  for all burn-ups considered. The main features distinguishing the RG versus WG fuels is the relative importance of  $^{239}\text{Pu}$  versus  $^{241}\text{Pu}$ . In the case of burning RG plutonium,  $^{241}\text{Pu}$  accounts for  $\sim 15\text{--}25\%$  of the fissions, with  $^{239}\text{Pu}$  representing a maximum 67% of the fissions.

*Partial MOX core loadings.* The fission fractions for loadings involving one-third MOX and two-thirds fresh LEU fuel are shown in Fig. 2. The fractions are quite distinct from those involving cores with 100% MOX fuel. For the partial MOX loadings,  $^{235}\text{U}$  and  $^{239}\text{Pu}$  are both dominant sources of fission. The two plutonium grades are most distinguishable by the relative importance of  $^{241}\text{Pu}$ , and by the value of the burn-up at which  $^{239}\text{Pu}$  overtakes  $^{235}\text{U}$  as the single largest contribution to the fissions. The fraction of fissions from  $^{238}\text{U}$  remains approximately constant throughout the burn and is 7–9% of the fissions.

### III. ANTINEUTRINO SIGNALS

Approximately 5–7 antineutrinos are emitted per fission, depending on the fissioning nucleus. These antineutrinos, that are produced in the beta decay of the fission fragments, all escape from the reactor and they cannot be screened. Their energies are in the range  $\sim 0\text{--}10$  MeV, and those with energies above 1.8 MeV can be detected by the inverse beta decay

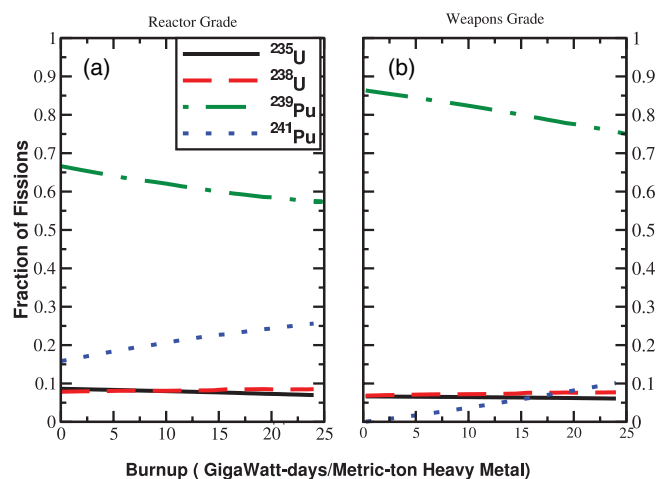


FIG. 1. (Color online) Fission fraction for  $^{235,238}\text{U}$  and  $^{239,241}\text{Pu}$  as a function of burn-up for pure MOX fuels. Panel (a) is for RG and (b) for WG plutonium. The two grades of Pu are distinguished by the relative importance of  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$ .

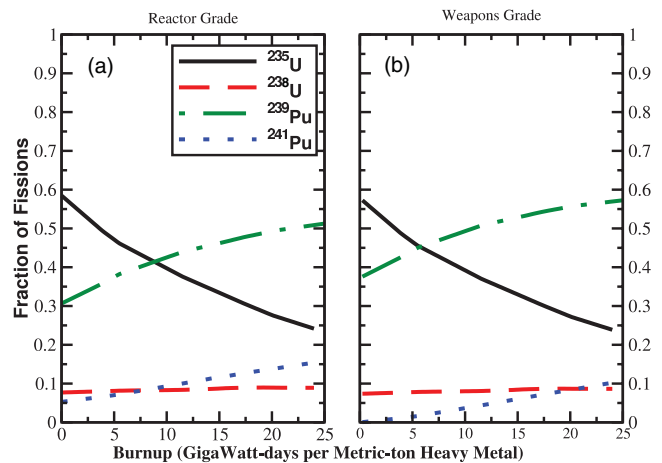


FIG. 2. (Color online) Fission fraction for  $^{235,238}\text{U}$  and  $^{239,241}\text{Pu}$  as a function of burn-up for 33.3% MOX and 66.7% LEU. Panel (a) is RG and (b) WG plutonium.

reaction on the proton ( $\bar{\nu}_e + p \rightarrow n + e^+$ ), with a cross section of the order of  $5 \times 10^{-43} \text{ cm}^2$  [1,6,7]. The differences in the fission fragment distributions for each actinide results in significant differences in the corresponding antineutrino spectra. We use the recently improved spectra of Mueller *et al.* [6] for all isotopes. Of course, the overall magnitude of the emitted antineutrino flux is controlled by the power density and the size of the core, and antineutrino safeguarding is only possible with independent knowledge of the reactor power.

The differences in the antineutrino signals for the different MOX loadings, can be displayed by introducing an *effective antineutrino detection cross section per fission* for each fuel.

$$\sigma_{\text{eff}}^j = \sum_i a_i^j \int \sigma(E_{\bar{\nu}}) S_i(E_{\bar{\nu}}) dE_{\bar{\nu}}, \quad (1)$$

where  $a_i^j$  is the fission fraction for fuel type  $j$  and each isotope  $i = ^{235}\text{U}, ^{238}\text{U}, ^{239}\text{Pu}, ^{241}\text{Pu}$ ,  $S_i(E)$  are the corresponding aggregate antineutrino spectra [6], and  $\sigma(E)$  is the cross

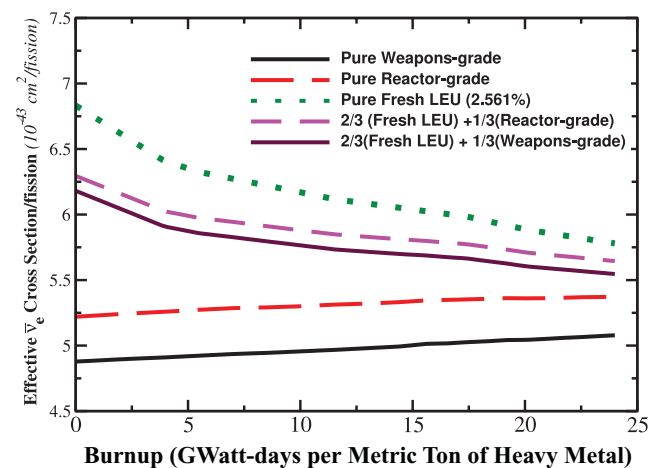


FIG. 3. (Color online) The effective antineutrino detection cross section per fission for each fuel type. For 100% MOX the effective cross sections *increase* with the burn, while for all loading with significant LEU they *decrease* with the burn.

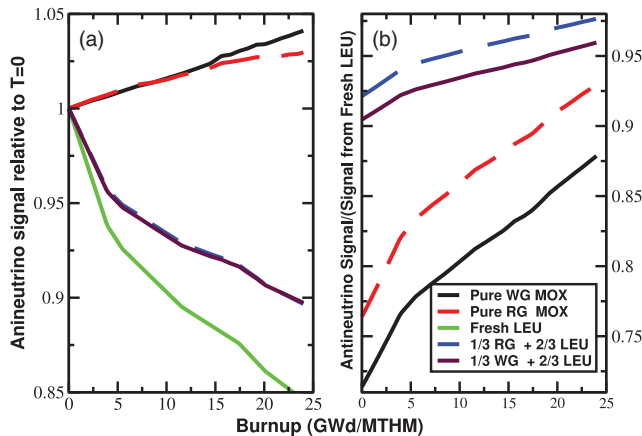


FIG. 4. (Color online) (a) The antineutrino signal relative to that at the start of the burn. (b) The ratio of the antineutrino signal for MOX fuels relative to that for fresh LEU.

section for antineutrino detection on the proton. The values of  $\int \sigma(E_{\bar{\nu}})S(E_{\bar{\nu}})dE_{\bar{\nu}}$  for each actinide, taken from [6], are listed in Table I. The *effective* cross sections of Eq. (1) vary with the burn-up for each fuel type as shown in Fig. 3, and it is clear that the MOX fuels are quite distinguishable from LEU. The information in Fig. 3 can also be displayed in terms of the change in the expected signal with the fuel burn-up relative to the signal at the beginning of the burn cycle. Alternatively, if a known fuel was monitored to calibrate the antineutrino detector, the ratio of the MOX signals to this known signal is useful. These different ratios are displayed in Fig. 4, where we take the “known” fuel to be fresh 2.56% LEU. As can be seen, pure and partial MOX fuel loadings are quite distinguishable from pure LEU. The larger the fraction the MOX fuel is of the reactor core, the more likely it is that the grade of plutonium can be deduced.

#### IV. SUMMARY

We investigate the antineutrino signals for four reactor core loadings of MOX; 100% MOX with RG or WG plutonium, and 33.3% MOX with RG or WG plutonium and 66.6% fresh LEU. In all cases we find that the antineutrino signals are quite distinct from that expected for pure LEU fuel burning with the same power density. The signals are distinguishable by the combination of their magnitudes and their rate of change

with fuel burn-up. In comparing the two different grades of plutonium, we find that their rate of change with respect to burn-up is similar, but that the magnitude of the signals for a given power density are different. This difference is maximum for reactor core loadings of 100% MOX, for which the RG signal is about 7% larger than that for WG. When the MOX represents one-third of the core, the signal difference between RG and WG is reduced to about 2%. If the thermal power is known, the overall uncertainty in the antineutrino flux emitted from the reactor is about 5%. The quoted [6,7] uncertainty in the number of antineutrinos per fission for  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Pu}$  is less than 3% and for  $^{238}\text{U}$  is 8%. When folded with the uncertainty in the thermal power measurement and the uncertainty in converting the thermal power to a fission rate, the total antineutrino flux is typically quoted with an accuracy of 3–5%. This overall uncertainty in the antineutrino flux, together with the calculations presented here, suggests that burning MOX plutonium fuels (within the class of MOX loadings considered) would be detectable using antineutrino monitoring. Distinguishing the grade of plutonium would be difficult unless the MOX represents the majority of the core.

Finally, we note that all of the calculations presented here are for fresh LEU and MOX fuels. In practice, a reactor core may be composed of fuel that is fresh, cycled through one year, two years, etc. The antineutrino signals for these will change depending on the core composition. Similarly, if less MOX plutonium is being burned than declared, the signal will change from that expected. The smoking gun for the latter scenario would be a slope for the variation in the antineutrino signal with burn-up relative to that for LEU that is flatter than the slopes for the MOX scenarios represented in Fig. 4(b). In general, the declared MOX content of the core would be most easily verified via antineutrino monitoring by taking a ratio of the observed antineutrino signal to a previously measured signal obtained by monitoring a fuel of known isotopic content.

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