Advances Towards Readily Deployable Antineutrino Detectors for Reactor Monitoring and Safeguards

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Abstract—The large flux of neutrinos that leaves a nuclear reactor carries information about two quantities of interest for safeguards: the reactor power and fissile inventory. Our SNL/LLNL collaboration has demonstrated that antineutrino-based nuclear reactor monitoring is feasible using a relatively small cubic scale detector made of Gadolinium loaded liquid scintillator at tens of meters standoff from a commercial Pressurized Water Reactor, deployed in an underground gallery that lies directly under the containment.

Recently we have investigated several technologies paths that could allow such devices to be more readily deployed in the field—of particular concern to reactor operators and safeguards practitioners is the flammability of the Gd doped liquid scintillator. In addition, many PWR facilities do not have an available underground gallery to provide the screening of muon induced backgrounds. As a result, we have developed and fielded three new detectors: a low cost, non-flammable water based design; a robust solid-state design based upon plastic scintillator; and a smaller cryogenic detector based on ultra-high purity Germanium. All three of these technologies have been deployed at our below-ground facility at the San Onofre Nuclear Generating Station in southern California.

We first present an overview of the use of antineutrinos in reactor monitoring. We then explain the detection mechanism based on inverse beta decay and the dominant sources of above-ground backgrounds that would contaminate this signal. Next, we discuss conceptual ideas under consideration for a future above-ground detector. Separate sections are devoted to describe the design, construction and deployment of each of our three new technologies that have already been deployment. We discuss the various levels of sensitivity to the reactor antineutrino signature that each of these detectors was able to demonstrate and the tradeoffs that accompany them.

Index Terms—Nuclear reactor safeguards, antineutrino detection, Cerenkov, HPGe.

I. INTRODUCTION

NUCLEAR reactors have served as the neutrino source for many fundamental physics experiments [1]. Conversely, the techniques developed by these experiments make it possible to use these very weakly interacting particles to monitor the power and fuel evolution of nuclear reactors [2]. The nature of the fuel monitoring is based on the difference in the antineutrino emission spectra from the main two fissile isotopes $^{235}\text{U}$ and $^{239}\text{Pu}$ ([3], [4]), and in the change in the isotope mass ratio in the reactor core during the fuel cycle (typically 1-1.5 years).

Several features of antineutrino detection are of interest to the International Atomic Energy Agency in its role as the international nuclear safeguards body. The highly penetrating nature of the particle and the high flux near reactors means that relatively small detectors external to the reactor can acquire a signal useful for monitoring, even with hour to day integration times. The technique provides real-time quantitative information about the reactor core power and isotopic compositions while the reactor is online. Moreover, the information about the detector status gathered with this method need not depend on operator declarations and can be kept under the control of the safeguards agency.

The field of antineutrino detection with small scale detectors is opportune reaching maturity at a time of resurgence of nuclear power. Even though this field has so far only found application in safeguarding one of the many elements of the nuclear fuel cycle (i.e., reactor safeguards), it is very important since it is at reactors where Pu is produced.

II. REACTOR MONITORING WITH ANTINEUTRINO DETECTORS

Antineutrino emission by nuclear reactors arises from the beta decay of neutron-rich fragments produced in heavy element fissions. On average, fission is followed by the production of approximately six antineutrinos. Since the energy spectrum of the produced antineutrinos differs significantly for the two major fissile isotopes $^{235}\text{U}$ and $^{239}\text{Pu}$, there is a corresponding difference in the average number of antineutrinos emitted per fission, $\Phi^{\nu}_i$. As the core evolves, $^{235}\text{U}$ is consumed and $^{239}\text{Pu}$ is produced by neutron capture on $^{238}\text{U}$. Hence, the core mass composition changes with the

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consequent change in fission rates \( N_i^f(t) \) from \(^{235}\text{U}\) and \(^{239}\text{Pu}\). The antineutrino emission rate also changes, according to the expression,

\[
n_\nu(t) = \sum_i N_i^f(t) \cdot \Phi_i^\nu
\]

(1)

where the sum is over the fissile isotopes.

In this paper, we will describe detectors that are based on two different antineutrino interactions: inverse beta-decay and neutrino coherent scattering. In general, the antineutrino detection rate \( N_\nu(t) \) for a given detector type depends on the interaction cross section \( \sigma_\nu(E_\nu) \) and the detector efficiency \( \varepsilon_\nu(E_\nu) \) through

\[
N_\nu(t) = \left( \frac{N_I}{4\pi D^2} \right) \sum_i N_i^f(t) \cdot \int dE_\nu \sigma_\nu E_\nu \varepsilon_\nu
\]

(2)

where \( \varepsilon_\nu(E_\nu) \) is the antineutrino energy-dependent number density per MeV and fission for the \( i \)th isotope, \( N_i \) is the number of interaction targets in the active volume of the detector, and \( D \) is the distance from the detector to the center of the reactor core.

Measuring \( N_\nu(t) \) does not provide enough information to determine the mass fraction of the fissile isotopes. Hence, the reactor thermal power \( P_{th} \), which is a known operational parameter, is used. The reactor thermal power is the rate of energy released in the reactor core as heat that is available for electricity production. Since antineutrinos emerge from the core isotropically and effectively without attenuation, they do not contribute to \( P_{th} \). However, the power contribution \( P_{th}^i \) from each fissile element is proportional to the isotope fission rate \( N_i^f \),

\[
P_{th} = \sum_i P_{th}^i(t) = \sum_i N_i^f(t) \cdot E_i^f,
\]

(3)

where \( E_i^f \) is the thermal energy released per fission per isotope. Therefore, by substituting (2) in (3), \( N_\nu(t) \) can be expressed in terms of the isotope fission rate ratio and of the thermal power \( P_{th} \). A useful regrouping of the terms in \( N_\nu(t) \) leads to

\[
N_\nu(t) = \gamma (1 + k(t)) P_{th}(t).
\]

(4)

Here, \( \gamma \) is a constant encompassing all non-varying terms chosen such that \( k \) is zero at the beginning of the reactor fuel cycle, and \( k(t) \) describes the change in the antineutrino flux due to changes in the reactor fuel composition. In the case when the thermal power is independent of the ratio of fission rates of each isotope, which is typical for most commercial reactors, temporal variations in \( N_\nu \) translate directly into variations in \( k \). Over the course of a reactor fuel cycle, \( k \) can decrease by 0.1, depending on the initial fuel loading, operating history and detector technology.

The relation between the mass fraction of fissile isotopes \(^{235}\text{U}\) and \(^{239}\text{Pu}\) and the detectable antineutrino flux is known as the burnup effect, and has been observed consistently in previous neutrino physics experiments [5]. The magnitude of this burnup effect can be predicted at the few percent level in an absolute sense, if the reactor fuel loading and the power history are known. Much of the uncertainty arises from systematic shifts in measured antineutrino energy densities \( \phi(E_\nu) \), so that the relative uncertainty in the predicted burnup rate can be considerably smaller.

III. TOWARDS AN ABOVE-GROUND ANTINEUTRINO DETECTOR

Our SNL/LLNL collaboration has demonstrated that antineutrino-based nuclear reactor monitoring is feasible using a relatively small cubic scale detector made of Gadolinium (Gd) loaded liquid scintillator at 25 meters standoff from commercial Pressurized Water Reactor (PWR) [2]. The deployment site was Unit 2 of the San Onofre Nuclear Generation Station (SONGS), which has maximum thermal (electric) power of 3.4GW\(_{th}\) (1.1GW\(_{elec}\)). With little or no burden on the plant operator we have been able to remotely and automatically monitor the reactor operational status (on/off), power level, and fuel burnup [6-8]. The detector was deployed in an underground gallery, the so-called tendon gallery, which lies directly under the containment dome of an operating PWR. The tendon gallery was located about 10m below the surface, providing \( \approx 25\text{m of water equivalent (mwe)} \) overburden which eliminates the soft component of the cosmic ray flux and reduces the muonic component by a factor of 7 [9].

A major and necessary step forward in antineutrino reactor monitoring is to demonstrate its feasibility for above-ground deployment. Many PWR facilities do not have an available underground gallery to provide the screening of muon induced backgrounds. Thus, having no overburden requirement in the detector design would vastly increase the range of possible field locations for safeguards applications. The current focus of our collaboration is to develop an above-ground detector able to report on/off reactor status. Further work will focus on developing a functional fully-sensitive reactor antineutrino detector.

The remainder of this section is devoted to explain the requirements that the signal-to-background ratio imposes on the detector design. In the following sections, the candidate technologies we are considering for an above-ground system are described. Some designs have already been tested at the reactor site and we explain the lessons learned from these initial deployments.
A. Signal from Inverse Beta-decay

Reactor antineutrinos are usually detected via the inverse beta-decay process on quasi-free protons in hydrogenous material. In this charged current interaction, the antineutrino $\bar{\nu}$ converts the proton into a neutron and a positron: $\bar{\nu} + p \rightarrow e^+ + n$. For this process, the cross section is small, with a numerical value of only $\sim 10^{-43}$ cm$^2$. The small cross section can be compensated for with an intense source such as a nuclear reactor. For example, cubic meter scale hydrogenous scintillator detectors, containing $\sim 10^{28}$ target protons $N_p$, will register thousands of interactions per day at standoff distances of 10-50 meters from typical commercial nuclear reactors.

The positron deposits energy via Bethe-Bloch ionization as it slows in the scintillator, and upon annihilation with an electron, emits two gamma rays which can deposit up to an additional 1.022 MeV of energy. Because the ionization and annihilation gamma ray energy depositions are effectively simultaneous, and promptly follow the antineutrino interaction, this combination of energy depositions is measured as a single event in the detector, referred to as the “prompt” energy deposition. Assuming complete containment of the event, the prompt energy and the antineutrino energy are directly related through the equation:

$$E_{\bar{\nu}} = E_{\text{prompt}} + 1.8\text{MeV} - 2m_e.$$  

The neutron carries away a few keV of energy from the antineutrino interaction and it is detected by capture after thermalization. A commonly used neutron capture agent is Gadolinium (Gd), due to its very large neutron capture cross-section. In liquid scintillator, 0.1% (by weight) Gd doping yields a neutron capture time of 28 µs and energy released of approximately 8 MeV via a gamma ray cascade. The interactions in the detector resulting from this 8 MeV cascade are referred to as the “delayed” energy deposition.

B. Background Rejection

For an antineutrino event, the positron $e^+$ and neutron $n$ are detected in close time coincidence, allowing strong rejection of the much more frequent singles backgrounds due to natural radioactivity or cosmic ray backgrounds. The incorporation of Gd in the detection material greatly aids this rejection by decreasing the neutron capture time to 28 µs, compared to a capture time of 200 µs for capture on hydrogen.

The time separation between prompt and delayed energy depositions follows an exponential distribution described by the neutron capture time. Unfortunately, this reaction is not the only means by which correlated pairs of interactions can be produced in the scintillator. For example, cosmic ray muons can produce fast neutrons in the material surrounding the detector – these neutrons can slow due to elastic collisions with protons, whose recoils are detected by the scintillator as a prompt signal, and then capture on Gd with the characteristic 28 µs time constant. In addition, the simultaneous arrival of multiple neutrons from a muon-induced shower could also produce several captures on Gd, whose relative timing would be distributed according to the 28 µs time constant. Such processes are referred to as correlated backgrounds.

The random coincidence of two unrelated single background interactions (gamma rays or neutrons) can also occur within the short time characteristic of neutron capture on Gd. The occurrence of these uncorrelated backgrounds is governed by Poisson statistics, and thus, their time separation follows an exponential distribution with time constant equal to the inverse of the single event rate (effectively the detector trigger rate).

C. Above-ground Background

An above-ground system based on the correlated detection of inverse beta-decay products would have to account for the increase in some components of the backgrounds, compared to a below-ground deployment. Gamma-ray rates depend on the surrounding amount of concrete and other radioactive materials specific to each site and not on the overburden depth. Cosmic-ray fluxes, on the other hand, are attenuated along the vertical depth and thus are larger on surface level locations. The net effect of the increase in cosmic-rays is to increase the overall neutron flux into the detector, fast and slow. The hadronic component, given by incident fast neutrons described by the Hess spectrum [10], is likely to dominate the correlated backgrounds.

Our collaboration is working on the development of a tight active and passive shield that will diminish the impact of these backgrounds. We are performing numerical simulations to investigate the effectiveness of shielding against the various neutrons backgrounds. It looks likely that significant reductions in the ambient thermal neutron rates can be easily accomplished; however the fast neutron backgrounds can likely only be attenuated by an order of magnitude. To survive in this environment, it appears that some sort of particle identification or insensitivity to this background will be required.

IV. PARTICLE IDENTIFICATION TECHNIQUES UNDER INVESTIGATION

The goal of particle identification is to reject signal-like background events. If, for example, the detector is designed to isolate neutron capture events, only event pairs where the neutron capture occurs second are accepted. Ideal background rejection would be achieved if both inverse beta-decay products are identified. Identification of other particles that largely contribute to background generation, like recoiling protons, can sufficiently improve the signal-to-noise ratio to achieve successful antineutrino detection.

One of the common methods for identifying fast-neutrons is through the use of Pulse Shape Discrimination (PSD). This method is based on the fact that certain organic scintillators demonstrate longer-lived excitations when interacting with heavier particles, such as protons. While not providing unambiguous identification of the positron, it allows separation
of gamma-like interactions from those of heavier particles and would thus allow rejection of prompt events from the primary correlated background due to fast-neutron interactions. However, liquid scintillators that offer good PSD are usually highly flammable and therefore, not desirable in most safeguards applications. In addition, it is difficult to maintain good PSD over long length-scales (greater than 50cm) and large volumes. As a result, using such a method for the scale needed for antineutrino detection would require the use of a large number of smaller detectors, leading to higher costs and complexities.

Our team is also investigating a new inorganic scintillator that allows PSD to be used to identify the delayed neutron capture. This material is known as LGB, where small grains of crystal $^6$Li$_{0.01}$Gd$_2$(BO$_3$)$_3$:Ce composed of the neutron capture agents Lithium, Gadolinium and Boron, are mixed with a plastic scintillator having a similar refractive index. We are currently studying neutron capture efficiency in this material where the PSD identifies the neutron capture on $^6$Li [11]. Practical factors against this choice of material for our detector design are the limited availability in shapes and sizes, and high cost. Of course, these factors can disappear if demand rises due to applicability in antineutrino detectors.

An alternative approach to unambiguously identifying the neutron capture event is to use a secondary neutron detector which is insensitive to gamma interactions, such as $^3$He or Li-glass. The previous materials being either prohibitively expensive or of limited availability, we have recently been exploring the use a $^6$Li-doped inorganic scintillator paint ($^6$Li mixed with ZnS(Ag)). The method integration with standard scintillator detectors and the readout of this material is still under development. However, there is an attraction to using such a material on the walls or septa of a segmented liquid scintillator detector since it could allow localization of the prompt and delayed events as a further method of background rejection.

Finally, another way to conceive particle identification was demonstrated by the Palo Verde experiment [12], using a highly segmented Gd-doped liquid scintillator detector. Since the positron annihilation will produced two gamma rays traveling in opposite directions that will likely deposit energy in more than one cell, contrary to electrons and protons that lose energy through ionizations in only one cell, multiplicity can be used for particle selection. In addition, the gamma-ray cascade from neutron capture in Gd (usually between 3-5 gammas) will also spread over several cells allowing for its identification. The drawback of a design of this sort is the possible low efficiency due to large amounts of dead material between cells and the high cost and complexity of instrumentation.

V. WATER CERENKOV DETECTOR

A water Cerenkov detector, doped with 0.1% Gd to facilitate the capture of neutrons produced in the inverse beta-decay process, has been built and deployed. In choosing to deploy a water Cerenkov detector, we were keenly aware of the low light output that results and hence low energy resolution. However, the potential advantage of such a detector is its insensitivity to the major correlated background of liquid scintillator detectors from fast neutrons. This is so because the recoiling protons due to fast neutrons scatterings have a very large threshold (> 2 GeV) for Cerenkov light production. In addition the inexpensive and inherently safe materials provide a significant advantage for real-world deployment.

A 250-liter tank of purified water, doped with 0.2% by weight GdCl$_3$, with no passive shielding was deployed in the tendon gallery at Unit 2 of SONGS. GdCl$_3$ dissolves in water and does not attenuate light in a detector of this size. A schematic of the detector is shown in Fig. 1. It consisted of a 250 liter UV transmitting acrylic tank filled with pure water and 0.2% GdCl$_3$. The outside walls of the tank were surrounded by diffusively reflective Tyvek.

![Figure 1: A schematic of the 250 liter prototype water Cerenkov detector.](image)

Before its deployment we tested and confirmed the detector’s sensitivity to detect correlated neutron events from a $^{252}$Cf source [13]. Since no passive shielding was used in this deployment, the uncorrelated background rate was fairly high (700 Hz). Interestingly, the correlated event rate due to pairs of neutrons generated at the same time (presumably by a single muon) was fairly high. It may, therefore, be possible in the future to reduce this background significantly with only a relatively thin layer of neutron capturing material, such as boron loaded polyethylene, because only one of the neutrons
needs to be stopped to eliminate a correlated pair. The results of this deployment are undergoing further analysis.

We are preparing a second deployment of a water Cerenkov detector in an above-ground location at SONGS, with an 800-liter tank of purified water similarly doped with 0.1% Gd and 10% photocathode coverage. This time, the detector will be inside the tight active and passive shield mentioned in section III.C. The shielding will have a minimum of 40 cm of surrounding polyethylene, inside which there will be an inch of borated polyethylene.

VI. PLASTIC SCINTILLATOR WITH GADOLINIUM LAYERS DETECTOR

The solid-state plastic scintillator detector described in this section is inherently robust and suitable for field deployment. The use of plastic scintillator means the device is non-flammable and non-toxic, can be fully assembled before deployment, and can be transported and deployed without special handling or hazardous material documentation.

This detector consists of 2-cm slabs of plastic scintillator (BC-408) interleaved with mylar sheets coated in Gd-loaded paint. Thus, the detection mechanism is identical to that previously described for the liquid scintillator. By using an inhomogeneous geometry, no compromise need be accepted in the light output, clarity, or timing properties of the plastic scintillator material over the ~1 meter length scale necessary for a practical antineutrino detector. On the other hand, this design contains a sizable amount of dead material in the main detector volume, and the proton number density is 10% smaller than in the liquid scintillator design. Moreover, the geometry reduces the neutron capture efficiency on Gd by ~25%.

As an initial test of the technology, 2 modules (approximately 300kg active material) were tested in the summer of 2007. They were deployed in the SONGS Unit-2 tendon gallery within the same experimental facility as the liquid scintillator detector described above. The two modules were installed within the existing neutron shield and directly replaced half of the liquid scintillator detectors, allowing a direct comparison of the performance of the plastic and liquid detectors.

These detectors were operated for almost a year and their performance was found to be about 10-20% worse than the liquid, in line with expectations. As shown in Fig. 2, an unplanned 6-day reactor outage was clearly visible in the measured rate. In addition, data was taken during a full reactor refueling outage and further analysis of that data will be forthcoming.

VII. GERMANIUM DETECTOR

A. Neutrino Coherent Scattering

Neutrino-nucleus coherent elastic scattering is a neutral current process predicted more than thirty years ago [14] but that has never been observed. The coherence of the scattering yields a cross section enhanced by the square of the number of components of the target. This $N^2$ enhancement, where $N$ is the number of neutrons of the target nucleus (the coefficient of the term proportional to the square of number of protons $Z^2$ is negligible), multiplies a factor of the order of $\sim 10^{-43}$ cm$^2$ in the expression for the cross section [15] for reactor antineutrino energies. In a target composed of heavy atoms (large $N$), an antineutrino reactor flux of about $10^{13}$ cm$^{-2}$ sec$^{-1}$ would produce a few hundred events per kg of detector material per day. It is the promise of such a dramatic reduction in the scale of a detector which makes this technology worth pursuing for an aboveground application. The reduction of a monitoring detector from 1 ton to 1-10 kg would provide an associated reduction in the interaction with cosmic-induced backgrounds. However, there will be additional backgrounds which are specific to the coherent process which must be considered.

The observation of neutrino coherent scattering requires the measurement of the energy of the recoiling nucleus. For a heavy nucleus, favored by a larger cross section, the recoil energy is proportional to the inverse of the nucleus mass $M$ with maximum value of few keVs for reactor antineutrino energies. Moreover, it is known that only a small portion of the kinetic energy of heavy atoms is converted into ionization of electrons. For example, the quenching effect reduces the ionization energy in Ge to 20% of the initial recoil energy [16], leaving only a few hundreds of electron-volts to detect. Therefore, a potential neutrino coherent scattering detector should be capable of isolating events with detectable energies as low as hundreds of electron-volts.

Given the small magnitude of the signal, very low detector operational thresholds are necessary to access reactor
antineutrino energies. A threshold of 300eV, for example, only gains us access to antineutrinos of ~8MeV or more. However, it is at the high end of their energy range that the reactor antineutrino spectra of \(^{235}\text{U}\) and \(^{239}\text{Pu}\) differ the most. For example, a Ge detector with threshold of 300 eV should record a decrease in the number of coherent scattering events in the course of the reactor fuel cycle of about 16%, whereas, with a 100-eV threshold it should register a 13% change. It is worth noticing that coherent scattering, if observed, would have sensitivity to variations in the number of antineutrino events slightly higher than inverse beta-decay.

Table I shows that, as the detector threshold decreases, the estimated number of counts per day for a 1-kg Ge detector rapidly increases. This is due to the increase in the number of detectable reactor antineutrinos as we gain access to lower incoming energies and to the increase in the number of above-threshold recoils for a given incoming energy. As an illustration of the relevance of detector threshold improvements, the last column estimates the Ge mass required for a three sigma confidence level in the difference in detected antineutrino events with 15-day measurements at the beginning and the end of the fuel cycle, without taking into account backgrounds. A 1-kg Ge detector with 100-eV threshold would do the job.

B. BEGe Detector and Deployment

The new prototype high-purity Ge detector, identified as Broad Energy Ge (BEGe) detector and developed by CANBERRA Industries in collaboration with the University of Chicago [16], operates at an energy threshold of 300 eV and a noise FWHM of 145eV. Several new features of its design allow for such unprecedented performance from a large-mass (~0.5 kg) Ge detector. These are: the crystal is a p-type which allows for better electron collection; the external n+ electrode is a lithium-drifted layer that provides shielding against low-energy betas and x-rays; the internal electrode is a point contact which reduces the crystal capacitance to ~ 1pF; and the new low-capacitance FET which delivers very low electronic noise. Other upgrades were made to reduce the radioactive backgrounds and the leakage current. Although this detector represents a remarkable advancement in Germanium detector technology, its performance level is still insufficient for a conclusive detection of neutrino coherent scattering.

Since December 2008, the BEGe detector has been deployed at the SONGS Unit 3 tendon gallery with the goal to characterize the location backgrounds with passive and active detector shielding. Thus, the system also comprises an anticoincidence Compton veto, an ultra-low background lead shield, 15 cm of polyethylene neutron shield, and internal and external muon veto paddles. One of the advantages of reactor monitoring with Germanium detectors is that the amount of detection material (number of Germanium crystals) can be increased without much of an increase in the system’s footprint.

Preliminary results from the first two months of data are shown in Fig. 3. This data included a 3 week period during which the reactor was off. As one can see, the detector performance is not yet sufficient to detect an antineutrino signature. However, progress has been made on both the reduction in low-energy backgrounds and the low-energy threshold. It should be noted that the backgrounds below 1 keV are now within a factor of 2 or 3 of what would be required for successful operation of a coherent scattering detector.

VIII. CONCLUSION

Antineutrino detectors will only be considered a viable technology for reactor safeguards once above-ground reactor power monitoring has been demonstrated. The main challenge for these detectors is how to deal with the increase in the cosmic-ray background at surface level. We have presented several ideas for detectors that either suppress or are insensitive to this dominant background. Besides our successful below-ground Gd-doped liquid scintillator, we have fielded three new detectors. These deployments have prepared the ground for new ones coming shortly. Although our initial goal is to establish reactor on/off status, our ultimate goal is to develop a functional above-ground detector fully sensitive to variations in reactor antineutrino signal.

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Fig. 3. Preliminary results form the first two months of data with BEGe detector. This data included a 3-week period during which the reactor was off.