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TOWARDS COMPACT ANTINEUTRINO DETECTORS FOR SAFEGUARDING NUCLEAR REACTORS

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Abstract.
In 2008 the IAEA Division of Technical Support convened a *Workshop on Antineutrino Detection for Safeguards Applications.* Two of the recommendations expressed that IAEA should consider antineutrino detection and monitoring in its current R&D program for safeguarding bulk-process reactors, and consider antineutrino detection and monitoring in its Safeguards by Design approaches for power and fissile inventory monitoring of new and next generation reactors. The workshop came to these recommendations after having assessed the results obtained at the San Onofre Nuclear Generator Station (SONGS) in California. A 600 litre, 10% efficiency detector, placed at 25m from the core was shown to record 300 net antineutrino events per day. The 2*2.5*2.5 m^3 footprint of the detector and the required below background operation, ~~prevents~~ an easy deployment at reactors. Moreover it does not provide spatial information of the fissile inventory and, because of the shape of a PBMR reactor, would not be representative for such type of reactor.

A solution to this drawback is to develop more efficient detectors that are less bulky and less sensitive to cosmic and natural radiation backgrounds. Antineutrino detection in the SONGS detector is based on the capture of antineutrinos by a proton resulting in a positron and neutron. In the SONGS detector the positron and neutron are detected by secondary gamma-rays. The efficiency of the SONGS detector is largely dominated by the low efficiency for gamma detection high background sensitivity

We are investigating two methods to resolve this problem, both leading to more compact detectors, which in a modular set up also will provide spatial information. One is based on detecting the positrons on their slowdown signal and the neutrons by capturing in ^{10}B or ^{6}Li, resulting in alpha-emission. The drawback for standard liquid scintillators doped with e.g. B is the low flame point of the solvent and the strong quenching of the alpha signal. Our research programs aim at developing scintillator materials, with a high flame point and weak or no quenching. Moreover we require (Pulse Shape Discrimination) PSD to differentiate between gammas and alphas and Bayesian filtering for background suppression. The second method is triggered by a recent paper indicating an effect of neutrinos on beta- decay. We are investigating the "mirror" reaction of antineutrinos on beta+ decay. Although we have reduced the effect already by two orders of magnitude, the present upper limit would still allow antineutrino-flux monitoring at nuclear power reactors.

1. Introduction

As stated in the introduction of the Final Report of the Focused Workshop on Antineutrino Detection for Safeguards Applications [1], "*Antineutrinos have unique features that make them especially interesting for IAEA safeguards: they cannot be shielded, are inextricable linked with the fission process, and provide direct real-time measurements of the operational status, power and fissile content of reactor cores, using equipment that is independent of reactor operation. Antineutrino detection offers a practical material* *accountancy capability for reactors. In this aspect it differs significantly from and is complementary to, the item accountancy, containment and surveillance measures which now prevail in the IAEA reactor safeguards regime.*" It should therefore no surprise that the recommendations of this workshop include that the IAEA should consider antineutrino monitoring as part of their Safeguards by Design approaches for power and fissile inventory monitoring of new and future generation reactors. But if this is so obvious, why are these monitors not yet installed? The answer is that antineutrino monitoring has only recently been demonstrated with a first-generation detector system, based on the technologies developed at large detector setups for scientific studies, mainly of antineutrino properties. The detector has a relatively low efficiency and hence is bulky due also to the necessary shielding.

In this paper we report on two supplementary ways to develop the second generation detector, which will be much more compact and easier to deploy.

2. State of the art

The most common way to detect antineutrinos is by making us of the so-called inverse β-decay in which an electron antineutrino is captured by a proton with a positron and a neutron as reaction products. This capture reaction is often presented as:

$$\overline{\nu_e} + p \rightarrow e^+ + n. \qquad (1)$$

Because the neutron is about 2000 times heavier than the positron, the laws of conservation of energy and momentum dictate that the positron mainly carries the energy information, whereas the neutron continues almost in the direction of the incoming antineutrino. The positron rapidly slows down and annihilates with an electron of the detection material and the two particles are converted into two γ-rays with an energy of 511 keV. These γ-rays transfer energy to the scintillation material by interactions with electrons. Neutrons are neutral particles and can only be detected indirectly. The most efficient way is through capture by a nucleus. As a result of this capture, γ-rays or secondary particles are emitted that will be detected. The capture reactions are usually resonances in a particular energy region and the neutron has to slow down by elastically scattering off (mainly) hydrogen nuclei to reach the energy, suitable for capture. Due to the size and energy difference of the positron and neutron the signal produced by the positron is earlier than signal produced by the neutron. This difference in time used to detect the two reaction products in delayed coincidence is the characteristic signature of having detected an antineutrino.

In general the state of the art detector systems use H or Gd as neutron capturers and also the neutron is identified by the γ-rays produced in the capture reaction of the neutron. Antineutrino signals therefore consist of two γ-ray like signals, detected in delayed coincidence. The γ-rays travel 10-30cm depending on material and energy, whereas the neutron travels of the order of 5cm [2, 3]. This implies that firstly all direction information carried by the neutron will be lost and secondly that a relatively large portion of the signal of neutrons detected near the wall will be lost, because they leave the detector undetected. These losses contribute to the low efficiency as reported for the detector tested at the San Onofre Nuclear Generating Station in California. For a segmented 600 litre detector an efficiency of about 10% was obtained [1, 4]. A consequence of such a low efficiency is that, to obtain sufficient statistics, the detector and its associated shielding becomes large, leading for the SONGS detector to a size of $2*2.5*2.5m^3$ and weighing several tonnes.

3. Alternative approaches

From the above it becomes clear that the neutron-capture probability and the detection efficiency for subsequent decay has a dominant impact on the detector dimensions. We are investigating two options to obtain compact antineutrino monitors:

- Developing and using detection materials in which the positron and neutron are detected in another mode other than by γ-rays;
- Investigating an indirect mode of detecting antineutrinos.

The two approaches will be discussed in the subsequent subsections.

3.1 Scintillation materials loaded with borate compounds

Nuclei like ^{6}Li and ^{10}B have low-energy resonances with high cross sections for (n,α) reactions. This implies that instead of γ-rays being detected, as by capture by H or Gd at thermal energies, the neutron is captured at higher energies and an α-particle is emitted. The Q-value for these (n,α) reactions is a few MeV, meaning that the energy of α-particles is in practice equal to the Q-value. The range of such α-particles in scintillation materials is a few microns. So the location of where the α-particles is stopped and produces light is the location of where the neutron was captured. By detecting the positron by its slow down scintillation, the positron range becomes of the order of a millimetre and hence the position of the positron and the neutron is well defined. If both positron and neutron is fully measured the dead volume of a detector is mainly determined by a dead layer at the end of the detector opposite to the side where the antineutrinos enter. This implies that e,g, for a cubical 40*40*40 cm^3 detector an efficiency of the order of 90% should be feasible.

This type of scintillation material has a number of advantages and disadvantages. Some of the scintillation liquids have a difference in pulse shape between signals produced by electrons (γ-rays or positrons) and signals originating from heavier charged particles like protons and α-particles. This so-called pulse-shape discrimination (PSD) helps to identify real antineutrino events from background. Moreover the resonances in ^{6}Li and ^{10}B occur at energies higher than the thermal energy and hence less neutron scattering will be required to reach the down to the resonance energy. This leads to a better conservation of the direction information, but also allows a shorter delayed coincidence time, which reduces background. For more details we refer to the simulations presented in ref. [2].

The scintillation materials also have distinct disadvantages. One of the most serious disadvantages are the energy quenching of the α-signal and the low flame point of the liquids. Up to recent the best boron doped liquid scintillation liquid had a flame point of -6 ˚C and the energy signal of the α-particle was quenched from 2.5 MeV to 60 keV, moving the signal from a clean part of the energy spectrum to the noisy low-energy part. Even with pulse-shape discrimination the signal is still prone to considerable background. Recently a new boron doped scintillation liquid was tested and showed in addition to pulse-shape discrimination a quenched α-particle energy peak at 110 keV. Moreover the flame point of the liquid was about 140 ˚C. The disadvantage of the new material relative to the old liquid is that in the new material only natural boron could be used.

The development of scintillation materials reached its maximum about 50 years ago. Especially in the last decade developments in material science have produced results that make the search for materials with better properties worth investigating. In our collaboration the development of novel liquid scintillator materials is performed by INCAS[3] together with the University of Groningen and industrial partners. The approach is based on loading liquid scintillators materials with trimethylborate [7] and optimizing the attenuation length, the stability and the solubility by the design of the surrounding ligands. The EARTH collaboration will design, build and provide test detectors. For the test of the developed materials we have a number of 5 cm diameter 10cm long cylindrical, 0.2 litre, cells with on both sides windows for light detectors. One cell filled with the high flame point boron doped scintillation liquid will be used as reference detector to compare with new developed materials. If a better material is found it will be used as reference detector.

A similar approach will be taken in the development of a Proof of Principle detector. Based on Monte Carlo simulations for light transport a detector was worked out and subsequently mechanically designed. We expect this detector with a volume of 36 litre and equipped with four PMTs to be constructed soon and to be filled with the high flame point, boron doped liquid scintillation material to be eventually installed at a nuclear power plant at about 20 m from the core. The main purpose of this detector is to optimise the characterisation of antineutrino signals, test the degree of directionality that can be obtained, and investigate the degree of background reduction that can be obtained by using active and passive shielding, and signal processing both on-and offline. Based on this experience a Proof of Principle detector will be designed, constructed, and tested. Needless to say that, technologies proven in the parallel development of scintillation materials and photon detection, will be implemented at suitable stages of development process.

3.2 Antineutrinos influencing β-decay

Presently there is a debate in the scientific literature on whether changes in the flux of solar neutrinos cause fluctuations in the decay rate of β^- emitters. This debate was triggered by the interpretations of Jenkins et al [5] of oscillations seen superimposed on top of the exponential decay as observed in the count rate of γ-rays following β^- decay of ^{32}Si, and of ^{226}Ra and progeny as well as γ-rays following β^- decay of ^{152}Eu,. The oscillations in the decay rate, have a magnitude of the order of 10^{-3}, have an oscillation period of one year, and extend over a period of several years. In addition a significant decrease in the count rate of γ-rays in the decay of ^{54}Mn by electron capture during, or prior to, solar flares at the end of December 2006. The reported effects are of the order of 10^{-4}. In their paper Jenkins et al attribute these to variations in the solar-neutrino flux as a result of the annual three percent variations in the Earth-Sun distance. Such flux variations are also put forward as the reason for the decrease in the ^{54}Mn γ-ray count rate during solar flares.

A number of papers, see reference [6] and references therein, challenge the interpretation by Jenkins and collaborators, and either explain the oscillations by temperature variations in the experimental set up or find no evidence for such oscillations related to the variations in the Earth-Sun distance. Experimentally the situation is therefore not yet clear.

For solar electron neutrinos, the flux at the Earth's surface is around $2*10^{10}$ $cm^{-2}s^{-1}$ and is very hard to vary. The situation for antineutrinos is quite different. One expects that if the effect is present for neutrinos it should also be observable for antineutrinos. The flux of antineutrinos coming from radiogenic processes in the Earth is estimated to be about 10^6 $cm^{-2}s^{-1}$. However, anthropogenic antineutrino sources are available in the form of nuclear reactors. A $1GW_{th}$ reactor produces about $3*10^{19}$ fissions per second (one fission yields ~$3*10^{-11}$ J). If the fission of uranium is exclusively of ^{235}U (6 antineutrinos per fission), the antineutrino source has a strength of about $2*10^{20}$ s^{-1}. Ignoring flavour-oscillation effects, at a distance of about 280m from the core the flux of antineutrinos is equivalent to the solar neutrino flux, and four orders of magnitude more than the flux of geoneutrinos. Under such conditions between reactor-on and reactor-off, temperature effects on the source-detector distance should be irrelevant. Larger antineutrino fluxes can always be obtained at closer distances to the reactor. If such an effect would be present the monitoring of nuclear power reactors would be feasible by looking for unexpected oscillations in the decay rate of β^+ emitters. And if the effect would be as large as claimed for the solar neutrinos this monitoring could be carried out from a distance of a few hundred metres from the reactor core.

To investigate if such an effect for antineutrinos is present for β^+ emitters, de Meijer et al [6] measured the decay rate of a number of β^+ and β^- emitters at 8m distance form the $2MW_{th}$ research reactor at the University of Technology Delft, the Netherlands, during subsequent periods of reactor-on, reactor-off and reactor-on. No effects have been observed for the two branches in the decay of ^{152}Eu, the decay of ^{137}Cs, ^{54}Mn and ^{22}Na. They derived an upper limit for the ratio $\Delta\lambda/\lambda$ for ^{22}Na, $(-1\pm2)*10^{-4}$, and for ^{54}Mn $(-1\pm4)*10^{-4}$. In comparison to the interpretation of Jenkins et al. [5] their measurements do not show any such effect to at least two orders of magnitude less. The authors conclude that either the hypothesis of Jenkins is not true or the effect of neutrinos on β^--decay differs considerably from the effect of antineutrinos on β^+-decay.

Although the result questions the effect of antineutrinos on β-decay, this possibility for reactor monitoring should not yet be fully discarded. The antineutrino flux at a distance of about 20 m from a 1 GW_{th} reactor will be about $4*10^{12}$ $cm^{-2}s^{-1}$; two orders of magnitude higher than in the Delft experiment. Again assuming a linear dependence on the antineutrino flux, the upper limit deduced from the Delft experiment, could still correspond to a 3% change in the decay constant. Such an effect could well be measurable and one may even expect that changes in fuel composition could be detected in this way at a distance of 20m. Such a measurement would therefore still be worthwhile and is presently in preparation.

4. Summary, discussion, conclusions and outlook

To study the properties of antineutrinos, large detector set-ups like KamLAND have been built. This large detector with a detector mass of one kiloton was based on the detection of the gamma radiation induced by the reaction products of antineutrino capture by a proton in the scintillation liquid. This type of detector has no direction sensitivity. For antineutrino monitoring of nuclear power stations, Bernstein, Bowden and collaborators [4,8] have constructed and tested a “down-scaled” version of the KamLAND detector for reactor monitoring. With this detector they proved, at the San Onofre Nuclear Generating Station, that it is possible to monitor the status of a power reactor and the changes in the fuel composition. Being based on the detection of gamma-rays, this detector has a low efficiency for antineutrino detection (about 10%) and hence has a large footprint.

We are investigating and developing compact and direction sensitive antineutrino detectors along two lines: directly, by the capture reaction of an antineutrino by a proton and indirectly, by investigating the change in count rate of gamma rays emitted by β^+ decay. In the former traditional mode, the capture reaction leads to a positron and a neutron. These two reaction products are identified by detecting the signal of the slowing down positron and the alpha-particle, following the capture of the neutron by ^{10}B. The range of the alpha-particle is a few micrometers, the range of the positron a few millimetres and of the neutron several centimetres. The neutron is mainly emitted in the direction of the incoming antineutrino. In this way the efficiency of the detector of 40*40*40 cm^3 is expected to be of the order of 90%. Moreover the detector has direction sensitivity and the traditional liquids allow for particle identification by pulse shape discrimination (PSD).

At present this type of detector has a number of severe drawbacks. Firstly the traditional scintillation liquids have a low flame point (-6 ˚C) and are classified as dangerous goods. Secondly the signal of the alpha-particle is heavily quenched meaning that the alpha particle with an energy of 2.5 MeV, shows up in the spectrum as a 60 keeV line.

Our development takes place along two parallel lines: designing, building and testing a 36 litre detector at a nuclear power plant and designing and testing new detector materials with a high flame point and a much smaller quenching. A first step has been the successful test of a Boron-loaded scintillation liquid with a flame point of 144 ˚C and an alpha-peak at 110 keeV. The liquid is not on the list of dangerous goods. We will fill our detector with this liquid and in parallel will continue to search for better detection materials.

In the indirect method we investigate if the effects of solar neutrinos on β^- decay, as claimed by Jenkins et al. [5] are also present for antineutrinos and β^+ decay. If the effect were the same for neutrinos and antineutrinos this method was expected to give a measurable effect at about 0.3 km from a 1 GW_e nuclear power plant. In a recent experiment [6] we have demonstrated that the effect is at least two orders of magnitude smaller for antineutrinos than claimed by Jenkins et al. for neutrinos. The present upper limit would however still allow monitoring nuclear power reactors at a distance of about 25m. The set-up can be made very compact and could be placed at several locations along the containment wall. In that way 3D tomography of a reactor would be feasible. This aspect is attractive enough to continue our investigation into the influence of antineutrinos on nuclear decay.

In summary, we would like to state that antineutrino monitoring of nuclear power reactors has been proven to be a very attractive tool to monitor the status and inventory of nuclear power plants. The next generation of detectors are likely to be much more efficient and hence more compact and will allow 3D imaging of the reactor core. This imaging is not only a great advantage for non-proliferation control, but also allows the operator of the reactor to improve the efficiency of the reactor in terms of fuel consumption.

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