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Paper Title

Quest for Very Compact Antineutrino Detectors for Safeguarding Nuclear Reactors

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Abstract:

Monitoring the status of a reactor and its fissile content in a continuous mode using antineutrinos is one of the challenging topics for safeguarding. Detectors deployed thus far require volumes of several cubic meters. In our search for a very compact antineutrino detector we have investigated an indirect method for antineutrino detection. In a first test of this hypothesis de Meijer et al. found an upper limit of $(\Delta\lambda/\lambda)=(-1\pm1)*10^{-4}$ at an antineutrino flux change of $2.5*10^{10}$ $cm^{-2}s^{-1}$ at the 2MW reactor at Delft, NL.
We continued our search for effects on β^+-decay at the nuclear power plant Koeberg, South Africa. Here we report on two sets of measurements, one in 2011, another from December 2012 - February 2014. In the first experiment a LaBr3 detector was used. La has a natural radioactive isotope, decaying by either β^- or EC, hence the effect of reactor-status change could be measured during background measurements: no effect was observed. With a ^{22}Na source an effect in the count rate was observed between reactor-OFF and reactor-ON which was not considered to be reliable, since an amplifier broke down and had to be replaced during the ramp-up of the reactor. In the recent measurement a 0.4L NaI detector coupled via a PMT to a PMT-base MCA was used. After overcoming a number of technical problems a stable condition has been reached. Again an effect has been observed in the count rate during two reactor changes. Provided that this effect is only due to antineutrinos affecting β^+-decay, this result would correspond to a change in decay constant $(\Delta\lambda/\lambda)= (-0.52\pm0.11)*10^{-4}$ at a flux change of $1.0*10^{13}$ $cm^{-2}s^{-1}$. We are in the process to investigate instrumental effects as alternative explanations.

Full paper:

1. Introduction

In nuclear reactors β^- emitting radionuclides are being produced, which in their decay emit antineutrinos. The intensity and energy spectrum of the antineutrinos depends on the rate of fuel consumption and the fissile composition. Since antineutrinos are hard to shield or to deflect, they will be present at the outside of the reactor containment. Measurements at the outside of the containment therefore present potentially a mean to continuously determine the status of the reactor and changes in the composition of the fuel. Hence antineutrino monitoring is of interest for safeguarding, and may be become a tool for instantaneous information of the "burn-up" process.

Thus far the preferred detection method for reactor antineutrinos is the so-called inverse β-decay in which an antineutrino is captured by a free proton and yields a positron and a neutron. The common way of detecting the positron and neutron is by the energy signal due to the energy loss of the annihilation radiation produced in scintillation materials by the positron and gamma-rays following the capture of the neutron. The energy spectrum of the antineutrinos peaks at ~0.5 MeV [1] and drops rapidly to about 8 MeV.

A drawbacks of this mode of detection is the threshold energy for this reaction of 1.8 MeV, which eliminates the majority of the antineutrinos to be detected. The low cross section for the inverse β-decay reaction and the large mean free path of the gamma-rays requires large volumes of scintillator material surrounded by extensive and heavy shielding. This has led to detectors at Rovno (former USSR), SONGS (California, USA) and NUCIFER (France) with volumes of more than 10 m^3, weighing ten tonnes or more. Moreover this mode of detection has no direction

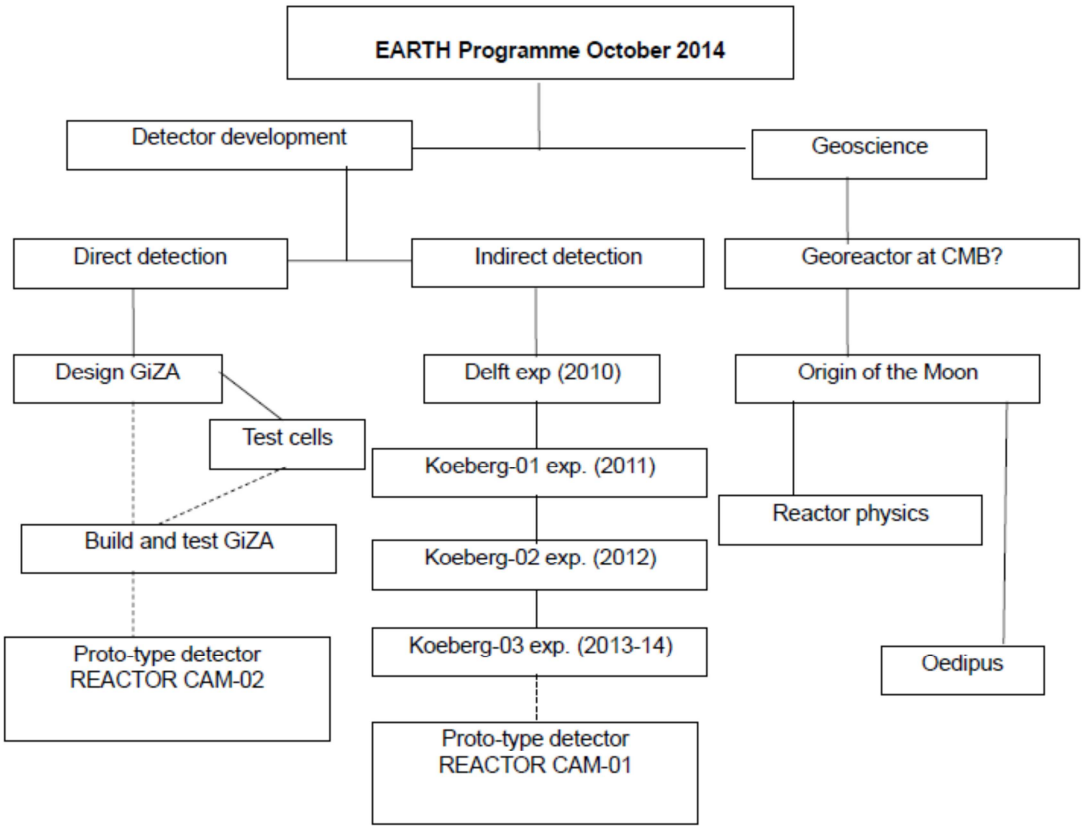

FIG.1. Status of the programme of Stichting EARTH at October 2014.

sensitivity, because the neutron is captured after it has thermalized and the gamma-rays are emitted isotropically.

In 2005 a foundation was established in the Netherlands: Stichting Earth AntineutRino TomograpHy (EARTH) to 3D-map the radiogenic heat sources in the Earth's interior by antineutrino tomography. From the onset it was realised that with existing technology this goal was out of reach and that newly-developed technologies had to be tested at the main source of antineutrinos namely nuclear power reactors. An overview of its program and its progress is depicted in Figure 1. The two main lines of research and development are Geoscience and Detector Development. The Geoscience part has addressed the feasibility of georeactors in the Earth's interior [2] and related to it proposed a hypothesis on the formation of the Moon [3].

The Detector Development part started with the development of a direction sensitive detector based on the inverse β-decay process, but with B-doped scintillator liquids. The nucleus ^{10}B has a low-energy resonance for capturing neutrons, thereby producing α-particles which only travel a few microns in the liquid. The positrons are detected by their slowdown, which is on the order of a few mm. This work led to the design of the detector GiZA. The further development was temporarily halted because of practical reasons. Testing such detectors at a nuclear power plant was not feasible due to the flammability and toxicity of the scintillation liquids. In a collaboration between INCAS[3], Assen NL) and the Stratingh Institute of Rijksuniverisiteit Groningen, NL, new scintillation materials such as B-loaded plastics and gels are being developed [4].

To further reduce the size of detectors an increase in cross section is imperative. Triggered by the work of Jenkins and Fischbach [5] we started to investigate the possibilities offered by indirect detection of antineutrinos with the original idea to rule out this mode of detection. The premise of this method is that antineutrinos may interact with a bound proton in a β^+ and electron capture (EC)-decaying radionucleus, which emits γ-radiation. The reaction with a bound proton has a positive Q-value and hence the method is sensitive to the entire antineutrino spectrum emitted by the reactor. This and the larger phase space enhance the sensitivity of detection by several orders of magnitude. In this reaction the antineutrinos would only affect the β^+-branch but not the EC-branch. This would lead to a change in decay constant, λ, and adjusted branching ratios. A first investigation of this method was carried out at the 2MW research reactor at Delft, NL. The measurements lead to an inconclusive result of $(\Delta\lambda/\lambda)=(-1\pm1)*10^{-4}$ at a flux change of $2.5*10^{10}$ $cm^{-2}s^{-1}$ [6].

In this paper we summarize the methodology and present data that indicate a possible effect. At this stage this effect is either due to a hidden instrumental effect or a physics process. In both cases the effect needs further measurements to identify the nature of the effect and to optimise the size of the effect and investigate whether this method leads indeed to a very Compact Antineutrino Monitor Reactor-CAM-01 as indicated in Figure 1.

2. Methodology

A set of ^{22}Na β^+-decay measurements were carried out at Koeberg Nuclear Power Station, ~30km northwest of Cape Town, South Africa in the period between 11 December 2013 and 9 May 2014. A scheduled power outage of the reactors was used to correlate count rate changes with changes of the antineutrino flux. For the details of the analytical methods applied and the assessment of systematic uncertainties we refer to a forthcoming paper [7]. In the present paper we describe the applied techniques, and present some of the results.

At the start of the measurements a ~1.3 kBq ^{22}Na source was placed inside the 51mm deep well of a 10.4 cm diameter, 10.4 cm long cylindrical NaI crystal. The detector was placed inside a 7.5 cm thick lead shielding with a copper intern lining. The detector is optically coupled via a quartz light guide to a 76 mm diameter low-K, Electron Tube 9305KB PMT which is surrounded by a solid mu metal shield and equipped with a 14 pins connector. This detector set-up was manufactured by SCIONIX–Holland: model 102 BP102 / 3M-X. It plugged into an ITECH Venus base. The Venus

is a universal digital multichannel analyser integrated into a photomultiplier base. It incorporates all the necessary electronics and was connected to a LENOVO laptop via the USB port. The on-

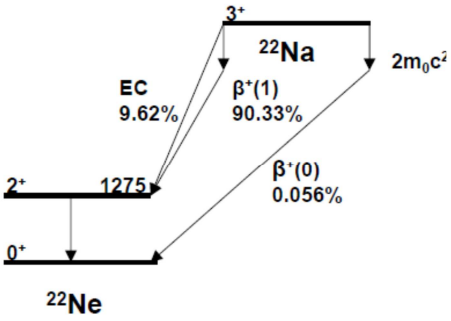

FIG. 2. Decay scheme of ^{22}Na.

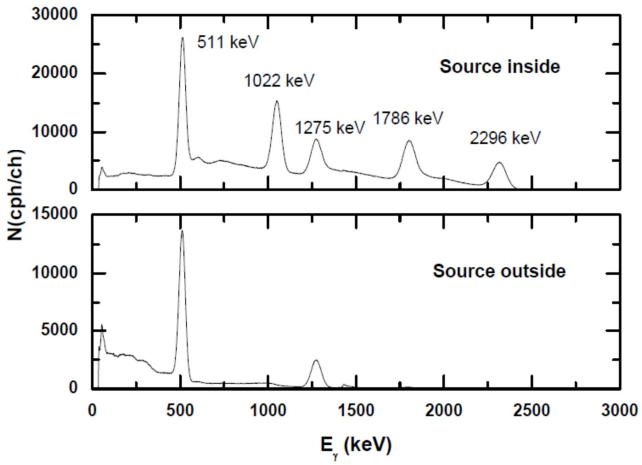

FIG.3. Gamma-ray spectra with the NaI well detector and the ^{22}Na source inside the well (top) and outside the detector (bottom).

line spectrum stabilisation and spectra
storage was controlled by the INTERWINNER software provided with the Venus base.

Figure 2 schematically shows the decay of ^{22}Na. The dominant decay proceeds through the first excited state of ^{22}Ne, leading to a 1275 keV γ-ray for EC and in addition, within the resolution of the detector, two annihilation quanta for the β^+-decay branch. The effect of coincidence summing is illustrated in Figure 3 where spectra with a ^{22}Na source inside and outside the well counter are presented. Please note that also the continuum part of the spectrum has changed shape and that continuum subtraction for net peak contents is not straightforward. From the figure it is clear that in the well counter events with E_γ>1275 keV are due to coincidence summing only. In this way the two decay branches are being observed more or less separately.

Gamma-ray spectra were collected for periods of two hours and stored in the memory of the laptop for further off-line analysis. In this analysis background was subtracted and the spectra were corrected for nuclear decay and some instrumental effects. A number of quality checks were made on a number of Regions Of Interest (ROI) to investigate the reliability of various analytical procedures. Three ROI's were considered to be of sufficient quality to be further analysed. This means that systematic uncertainties for these ROIs were small compared to the statistical ones. These ROI's are:

- Gate Total: 169< E_γ<2451 keV, compassing almost the entire spectrum;
- Gate 1275: 1152< E_γ <1351 keV, containing the E_γ=1275 keV peak;
- Gate I: 1353<E_γ<1921 keV, a broad gate over the summing part of the spectrum due to β^+-decay only.

3. Results

After correcting for decay and some instrumental effects the analysis of the data was carried out for a total of two ON-OFF-ON transitions of reactor unit#1: one in February-May 2013 and a second one in the period October 2013-February 2014. This resulted in the two data sets for the three ROIs. For each sub-period (ON or OFF) the weighted average of the count rate and its statistical uncertainty in each gate were determined. The results are listed in Table 1. The data in Table 1 show that the two ON-OFF-ON periods each lasted about 100 days, so the counts rates per

sub-period are averaged over about 300 to 400 two-hour spectra. The table lists the relative change in count rate. One notices that for the ROIs Gate Total and Gate I the ΔA/A values are negative whereas for Gate 1275, the value is positive. Moreover this pattern is present in the both periods and therefore reproducible. As one notices the combined effect (in bold) for both Gate Total and Gate I is statistically significant by almost 5σ.

Thus far only statistical uncertainties have been included but with the high precision data systematic error and uncertainties may play a more significant role. In our forthcoming paper [7] the contributions due to analytical procedures, pile-up and baseline restoring, background and antineutrino flux have been assessed. For the potentially most significant contribution to systematic uncertainties the analytical procedures were assessed by calculating count rate ratios of Gate 1275/Gate I in three ways:

1. As a direct ratio for every data point without corrections for the decay of the source and instrumental effects. (No-DIC).
2. The direct ratio for every data point after correction for decay and instrumental effects (DIC)
3. The ratio of the weighted averages for each of the reactor-ON/OFF periods. This method is indicated as RWA.

Table 1. Value of the average difference between ON and OFF count rates. The uncertainties represent 1σ values.

DOY 2013	Status	$\left[\frac{\Delta A}{A_{OFF}}\right] * 10^4$ Total	$\left[\frac{\Delta A}{A_{OFF}}\right] * 10^4$ 1275	$\left[\frac{\Delta A}{A_{OFF}}\right] * 10^4$ Gate I
53-80	ON			
87-112	OFF	-0.66±0.16	1.4±0.5	-2.3±0.3
122-140	ON			
		-0.52±0.11	***0.9±0.5***	***-1.9±0.4***
282-315	ON			
323-362	OFF	-0.39±0.16	0.4±0.5	-1.6±0.3
368-389	ON			

Table 2. Values of the count rate ratio R for three analytical procedures for two periods with a reactor-ON-OFF-ON sequence.

DOY 2013	Status	R_1 No-DIC	R_2 DIC	R_3 RWA	$\Delta R_1 * 10^4$	$\Delta R_2 * 10^4$	$\Delta R_3 * 10^4$
53-80	ON	0.51503(3)	0.51220(3)	0.51239(4)	1.6±0.5	1.7±0.4	1.5±0.5
87-112	OFF	0.51487(3)	0.51204(3)	0.51224(3)	***1.8±0.3***	***1.9±0.3***	***1.9±0.3***
122-140	ON	0.51507(4)	0.51225(4)	0.51246(4)	2.0±0.5	2.1±0.5	2.2±0.5
282-315	ON	0.51016(3)	0.51200(3)	0.51213(3)	1.0±0.4	1.2±0.4	1.1±0.4
323-362	OFF	0.51006(3)	0.51188(3)	0.51202(3)	***0.9±0.4***	***1.0±0.4***	***1.0±0.3***
368-389	ON	0.51014(3)	0.51197(3)	0.51211(3)	0.8±0.4	0.9±0.4	1.0±0.4

The first method contains the least assumptions and is based on the fact that common factors in numerator and denominator cancel in a ratio. These common factors include source strength and live time . In the second method the assumption is made that the instrumental effect could be

corrected for. The corrections for decay (source strength) and live time are the same for the corresponding data points and will drop out in the ratio. In the third method the instrumental effects and decay correction and variations in live time end up in the weighted average, possibly after a linear fit to the data set of the three reactor-status periods and subsequently pivoted. This method contains the largest systematic uncertainty. The three methods are therefore listed in order of an increasing number of assumptions for details see ref.[7].

Table 2 presents a comparison of the results of the three methods during two periods in which the reactor status switched from ON-OFF-ON. For each of the periods there is no significant difference in the change in count rate ratio, ΔR, between the three analytical methods despite small differences between the corresponding R-values. The ΔR values for the two periods differ by a factor of two. Two possible explanations are: effects caused by the insufficient pile-up and/or base-line restoration were only partly compensated for by the linear spectrum stabilisation, and/or there was a change over time in the reactor-fuel composition.
From Table 2 we conclude that the contribution from the analytical procedures to the total uncertainty is not significant compared to the statistical uncertainties.

4. Acknowledgements

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Conclusions:

The goal of this investigation was to find out if changes in reactor status would lead to changes in count rate of gamma-rays emitted by a ^{22}Na source and if these changes could be attributed to changes in antineutrino flux and hence if the effect could be the basis for a very compact antineutrino monitor for safeguarding purposes.

This work shows that there is a statistically significant (almost 5σ) effect in the relative count rate between reactor-ON and -OFF. Moreover the effect has been observed in two consecutive ON-OFF-ON periods. The effect shows for reactor-ON a drop in count rate of about 10^{-4} for a ROI on the almost entire γ-ray spectrum and for a ROI containing counts only due to β^+-decay. For a ROI in which both electron capture and β^+-decay contribute, there is an increase in count rate.

Various possible contributions to systematic errors and uncertainties were assessed and it was concluded that these contributions to the uncertainty in the results was small compared to the

analytical uncertainties. An instrumental effect that has escaped our attention should explain why the count rate drops for the ROI on almost the entire spectrum and the ROI Gate I on β^+-events only and increases for its adjacent ROI Gate 1275.

The above observations are consistent with antineutrinos affecting the β^+-decay of ^{22}Na and leaving EC unaffected. It would mean that the overall decay constant is reduced by a factor $(0.51\pm0.11)*10^{-4}$ and that the branching ratio for β^+-decay has been reduced by a factor $(1.9\pm0.4)*10^{-4}$ in favour of EC.

The change in total antineutrino flux is calculated to be of $1.48*10^{13}$ $cm^{-2}s^{-1}$ at the position of the ^{22}Na source. In case the effect is due to antineutrinos interacting with ^{22}Na, the cross section for the reaction is $(2.9\pm0.6)*10^{-26}$ cm^2 with in addition a systematic uncertainty of about 15%. This value is about 15 orders of magnitude larger than extrapolated from the capture cross section on a free proton and taking into account the larger phase space and the fact that ^{22}Na contains 11 protons. From the point of view of safeguarding the reason for the effect is of course of interest because it determines in what way the effect can be optimised to the extent that it can be used as a compact antineutrino monitor. A number of additional measurements are planned to better identify the cause of the effect and develop it to a proto-type of Reactor-CAM-01, as indicated in Figure. 1.

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