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### **Reactor Neutrino Detection for Non Proliferation with the NUCIFER Experiment**

Th. Lasserre, V.M. Bui, M. Cribier, A. Cucoanes, M. Fallot, M. Fechner, J. Gaffiot, L. Giot, R. Granelli, A. Letourneau, D. Lhuillier, J. Martino, G. Mention, D. Motta, Th.A. Mueller, A. Porta, R. Queval, J. L. Sida, C. Varignon, F. Yermia

Corresponding author: thierry.lasserre@cea.fr

Neutrinos are the most abundant matter particles in the Universe. Thoroughly investigated in basic science, the neutrino field is now delivering first applications for nuclear reactor monitoring. We present here the NUCIFER neutrino experiment to automatically and non-intrusively monitor nuclear power plant thermal power and Plutonium content. The core of the detector is a one ton Gadolinium-doped liquid scintillator tank to be installed in a basement room less than 30 m from a reactor core. The Division of Technical Support (SGTS) within the IAEA Department of Safeguards is currently investigating the potentiality of neutrinos as a novel safeguards tool.

### 1. Neutrino production and detection at nuclear reactors

Nuclear reactor power comes from the energy produced by the fission of heavy elements (i.e. U and Pu) into neutron rich nuclei. Reactor neutrinos are produced by the  $\beta$ -decay of these fission products (FPs) into more stable nuclei:  ${}^{A}_{Z}X \rightarrow {}^{A}_{Z-1}Y + e^{-} + \bar{v_{e}}$ . Each fission releases about 200 MeV and 6 antineutrinos, which means that the flux emitted by a 1 GW<sub>th</sub> reactor is ~1.5 10<sup>20</sup> antineutrinos/second. Although the interaction cross section between matter and neutrinos is very tiny (~10<sup>-43</sup> cm<sup>2</sup>), the huge emitted flux allows us to detect their signal with a relatively small detector (3 m x 3 m) located a few tens of meter from the core.

In NUCIFER reactor antineutrinos are detected in liquid scintillator doped with Gadolinium (Gd). The detection reaction is the inverse  $\beta$ -decay  $v_e + p \rightarrow e^+ + n$  (1.8 MeV threshold). The e<sup>+</sup> produces a prompt energy deposition carrying the neutrino energy. This first signal is followed by a delayed energy deposition induced by the radiative capture of the neutron on Gd with the emission of a gamma cascade of total energy 8MeV. Since the number of emitted neutrinos and their mean energy depend on the fissioning isotopes (<sup>235,238</sup>U, <sup>239,421</sup>Pu), their detection provides a direct image of the core composition.

# 2. Sensitivity to the fuel composition

The two main fissile isotopes contained in the fuel of a pressurized water reactor (PWR) are <sup>235</sup>U and <sup>239</sup>Pu. Fresh uranium fuel is typically enriched at 3.5% in <sup>235</sup>U, while <sup>239</sup>Pu is produced by neutron captures on the original <sup>238</sup>U followed by two consecutive  $\beta$ -decays: <sup>239</sup>U $\rightarrow$ <sup>239</sup>Np $\rightarrow$ <sup>239</sup>Pu. During a reactor cycle <sup>235</sup>U is burned while the net effect on the <sup>239</sup>Pu quantity is an accumulation. This means that the relative contribution to the total number of fissions induced by these two isotopes changes over time: it increases for the <sup>239</sup>Pu while decreasing for the <sup>235</sup>U. This is called the "burn-up" effect.

Fission parameters	<sup>235</sup> U	<sup>239</sup> Pu
Energy per fission (MeV)	193.5	198.9
Mean detectable $\nu$ energy (MeV)	2.94	2.84
Detectable v number (per fission)	1.92	1.45
Cross section $<\sigma_{int}>$ (10 <sup>-43</sup> cm <sup>2</sup> )	3.20	2.76

Table 1. Relevant fission parameters of <sup>235</sup>U and <sup>239</sup>Pu

At the end of a cycle, both isotopes basically share half of the emitted power. Because the fission products of these two isotopes have different atomic masses, their  $\beta$ -decays produce different neutrino fluxes with different energy spectra. The key parameters of <sup>235</sup>U and <sup>239</sup>Pu fissions are summarized in Table 1. This feature is at the origin of the sensitivity of the antineutrino probe to the plutonium content of the core. For

instance if we consider the hypothetic case where all fissions would come from pure <sup>235</sup>U or pure <sup>239</sup>Pu to produce the same thermal power, the ratio of detected antineutrinos would be:

$$\frac{N_{v}^{U}}{N_{v}^{Pu}} = \frac{\left(\frac{N_{v}}{\text{fission}} \times \sigma\right)^{U}}{\left(\frac{N_{v}}{\text{fission}} \times \sigma\right)^{Pu}} \frac{\left(\frac{E}{\text{fission}}\right)^{Pu}}{\left(\frac{E}{\text{fission}}\right)^{U}} \approx 1.6$$

This large difference suggests the possibility to use the antineutrino rate to monitor changes in the relative amounts of <sup>235</sup>U and <sup>239</sup>Pu in the core, with some underlying difficulties to be discussed later. Smaller contributions of the two other fissile isotopes (<sup>238</sup>U and <sup>241</sup>Pu) are taken into account in the simulation and analysis.

#### 3. Sensitivity to Thermal Power

The thermal power  $P_{th}$  of a reactor core can be calculated from the detected neutrino flux  $N_v$  by using the formula:

$$N_v = \gamma (1 + k(t)) P_{th}$$

where  $\gamma$  is a constant proportionality factor including the target mass, detection efficiency, solid angle, etc. and (1+k(t)) is a time dependent factor which takes into account the change in fuel composition (i.e. in neutrino flux) induced by the "burn-up" effect described in the previous section. Knowing the initial fuel composition and simulating its evolution over the reactor cycle we can calculate this factor. Its effect over one typical cycle of a PWR is of the order of 10%.

The interest of power measurement through neutrino flux detection is double: it can provide a cross-check of the already existing method of power monitoring and, by moving the small neutrino detector to different power plants, we can have the unique opportunity of cross-calibrating different reactor types or cross-calibrating the same type of reactor at different sites. This novel idea is being developed in the NUCIFER collaboration.

### 4. The NUCIFER detector

The detector has been designed according to the IAEA division of Safeguards Technical Support (SGTS) recommendations [1]. The detector is small compared to neutrino detection standards, relocable, remote controlled, safe, and temper-proofed [2]. To optimize and validate the detector and shielding geometry we developed a dedicated Monte-Carlo simulation and we performed background measurement campaigns at the two research reactors where NUCIFER will be first tested [3]. The resulting detector (**Figure 1**) is composed of a stainless steel cylindrical tank (height 1.7 m, diameter 1.2 m) filled with 0.85 m<sup>3</sup> of Gd-doped liquid scintillator. 16 photomultiplier tubes observe the active volume from the top through a 25 cm thick acrylic light guide ensuring uniform response of the active volume and improving safety requirements. For calibration purposes, a LED based light injection system allows to correct for regular instrumental drifts. In addition small radioactive sources could be deployed along the target central axis inside a vertical tube. From inner to outer layers the inner detector vessel is surrounded by an active veto made of 5 cm thick plastic scintillator panels, 14 cm of Polyethylene, and 10 cm of Lead. The veto will tag the passage of cosmic ray muons close to the detector and will suspend data acquisition in order to supress induced neutron background [4]. It must be deployed at least a few meters underground to reduce the cosmic ray induced backgrounds.



Figure 1: The NUCIFER detector vessel surrounded by its cosmic ray muon veto and passive lead and polyethylene shielding. The active volume is composed of 850 liters of Gadolinium doped liquid scintillator.

#### 5. Current Status

All detectors components have been procured in 2009-10 and the (unshielded) detector has been taking calibration data for 6 months in order to fully commission the data acquisition system. We used an unloaded liquid scintillator composed of linear alkyl benzene (LAB) with a fluor (PPO, 2 g/l) and a wavelength shifter (bis-MSB, 20 mg/l). Radioactive sources of Americium-Berylium, Cesium 137, Cobalt 60 and Sodium 22 have been deployed in a Teflon coated stainless steel tube reaching the center of the active volume. Results of the comparison measured and simulated energy scales are displayed in **Figure 2**. They show a excellent agreement in the region of antineutrino energy between 667 keV and 5.5 MeV, within a few percent. This attests for the high-quality detector performances as well as for the goodunderstanding of the detector response that is mandatory for the selection of the neutrino candidates at reactors.



Figure 2: Results of the calibration of the energy reconstruction of gamma ray and neutron events in the unshielded NUCIFER detector (blue) compared with the simulation (red).

However neutrino selection will be also based on the selection of two energy depositions in coincidence within 100 μs. We thus performed a calibration run using an Am-Be radioactive source emitting neutrons in coincidence with a 4.4 MeV gammas, with a rate of 90 Bq.



**Figure 3** shows the distribution of the delay time between the prompt 4.4 MeV gamma and the delayed neutron candidates. Results can easily be fitted with a sum of an exponential and a constant, the first with a 209  $\mu$ s time constant attesting clearly the neutron capture on hydrogen, and the second being consistent with the expected accidental background of our detector (naked vessel without lead shielding).



Figure 3: Results of neutron calibration of the unshielded NUCIFER detector using an Am-Be radioactive source. The plot displays the time delay between the prompt 4.4 MeV gamma ray and candidates of a neutron capture on Hydrogen. A clear exponential decay with a 209 µs time constant (red dashed curve) appears on top of the flat uncorrelated background (blue dotted line), attesting for the neutron capture signal in our unloaded liquid scintillator (LAB).

### 6. Deployment schedule

Presently we are installing the detector 7 m away from the Saclay-Osiris research reactor core at the French Atomic Energy and Alternative Energies Commission. Starting at the end of 2010 this first phase at the world nearest distance from a nuclear core is foreseen to last one year. In 2012 the detector should be moved at the ILL research reactor of Grenoble. Thanks to its almost pure <sup>235</sup>U fuel composition, versus 20% <sup>235</sup>U enrichment at Osiris, we will be able to perform a precise characterization of the detector thanks to the very well known fuel composition. In 2013 we plan to deploy NUCIFER close to a commercial nuclear reactor, possibly in a country under Safeguards to finally demonstrate the potential of the NUCIFER concept, possibly in collaboration with the IAEA.

### 7. Nuclear fuel online monitoring

The detailed GEANT4 simulation of Nucifer developed to optimize its design has also been used to study its performance and its response to reactor neutrinos. The estimated energy resolution for the detection of 4

MeV gammas randomly generated in the target is 20% while the estimated reactor neutrino detection efficiency is 50%. Beside the detector simulation a detailed simulation of reactor neutrino emission has been performed. It consists of a dedicated twofold code: the MCNP Utility Reactor Evolution (MURE) package, developed by the CNRS/IN2P3 laboratories LPSC Grenoble and IPN Orsay [5] is used for realistic reactor core simulations. MURE is a precision code written in C++ which automates the preparation and computation of successive MCNP (Monte-Carlo N-Particle) [6] calculations either for precision burn-up or thermal-hydraulics purpose. The MURE outputs feed a second part, called BESTIOLE [7], which incorporates precise measurements of electron spectra at ILL [8] as well as updated nuclear databases to predict the spectra per fission of each isotope. Merging the results from this simulation to the detector response we have been able to reproduce different scenarios of neutrino detected rate corresponding to different fuel compositions presented below. More complex diversion scenarios associated to several reactor concepts have been performed and are presented in [9].

Figure 4 shows the expected weekly-detected rate of neutrinos generated by a 2.9 GW<sub>th</sub> PWR placed 25 meters from NUCIFER. At the beginning of the cycle the fuel composition in relative number of fissions of  $^{235}\text{U}/^{239}\text{Pu}$  is 70/30%, while it is 50/50% at the end of the cycle. We can clearly see the decrease in the neutrino rate induced by the burn-up effect. The gap after week 51 simulates reactor stop to refuel the core. After week 67 the reactor starts again with the initial fuel composition. The relative neutrino rate change is ~7%, produced by a 20% relative change in the number of fissions induced by  $^{235}\text{U}$  and  $^{239}\text{Pu}$ .



Figure 4: NUCIFER performance monitoring the thermal power and burnup of a 2.9 GW<sub>th</sub> PWR located 25 m away. Each data point represents the weekly neutrino rate detected. During week 0 the core composition in relative number of fissions of <sup>235</sup>U and <sup>239</sup>Pu is 70/30%, while it is 50/50% at the end of the cycle (week 50). The observation of the decrease in the neutrino rate allows monitoring the Pu content in real time. The period between weeks 51 and 67 simulates a reactor outage with fresh fuel replacement. Starting at the same thermal power for the study, the relative neutrino rate change is ~7% after week 67, induced by a 20% relative change in the number of  $^{235}$ U and  $^{239}$ Pu fissions.

#### 8. Sensitivity to illicit Plutonium retrievals from a nuclear reactor core

In this section we address the sensitivity of the NUCIFER detector in discovering illicit Plutonium retrieval for a PWR of 2.9 GW<sub>th</sub> located 25 m away from the detector. We compare the detected neutrino rates before and after the reactor stop by fixing a constant reference composition of 70/30% in the number of fissions induced respectively by <sup>235</sup>U and <sup>239</sup>Pu after the stop and by varying the compositions before the stop with the following values: 65/35%, 62.5/36.7%, 60/40%, 55/45% and 50/50%. Each different composition before the stop corresponds to a certain mass of Plutonium produced during the cycle that can be extracted during the stop. This mass can be calculated from the composition by knowing the thermal power (P<sub>th</sub> = 2.9 GW<sub>th</sub>), the cross section for neutron induced fission ( $\sigma_{(f,n)} = 120$  b) and the neutron flux ( $\phi_n = 3.5 \ 10^{14}$  n cm<sup>-2</sup> s<sup>-1</sup>). We calculated that the 5 considered compositions before the stop correspond to the following masses of extracted Plutonium: 55, 80, 105, 155 and 190 kg. Each different composition before the stop also corresponds to a different detected neutrino rate, while the neutrino rate after the stop is fixed by the reference composition. To calculate our sensitivity to Plutonium retrieval we use the  $\chi^2$  distribution method. Depending on the quantity of Plutonium retrieved during the reactor stop, the  $\chi^2$  of the two measurements

will follow a specific  $\chi^2$  distribution. Thus for a given value of  $\chi^2$  we can calculate the probability of issuing false alarms as a function of the probability of issuing valid alarms for the retrieval of a selected Plutonium mass.

**Figure 5** shows the curves obtained for various Plutonium masses in the plane of the two above probabilities by varying the  $\chi^2$  value. If we require an upper limit on the probability of false alarms of 4% and a lower limit for the probability of valid alarms of 75% we select the blue zone. The fact that the curves for mass  $\geq 80$  kg cross this area means that, with the required probabilities of valid and false alarms, we are sensitive up to this quantity of extracted Plutonium. This result is for a statistics of the two relative neutrino flux measurements corresponding to 15 days of data taking with our detector placed at 25 m from the considered 2.9 GW<sub>th</sub> PWR. Different reactor powers, data taking periods and detector distances would lead to different sensitivities.



Figure 5: NUCIFER sensitivity to illicit Plutonium retrievals from a nuclear reactor core. The figure shows the distribution of the probability of issuing false alarms as a function of the probability of issuing valid alarms for the retrieval of a certain Pu mass. Requiring a probability of false alarms  $\leq 4\%$  and a probability of valid alarms  $\geq 75\%$  NUCIFER is sensitive a diversion of a Plutonium mass  $\geq 80$  kg, considering two relative measurements with a statistics of 15 days each before and after the diversion.

# 9. Conclusions

We review the status and potential NUCIFER neutrino experiment aiming to demonstrate the possibility of high accuracy, reliable, and temper-proof monitoring of fission nuclear reactor thermal power and detecting undeclared Plutonium retrieval. The detector has been tested in an almost final configuration and calibration preliminary results indicate a good understanding of the detector time and energy responses. This attest for the readiness of NUCIFER for the reactor antineutrino hunt. The detector is currently being integrated at the OSIRIS research reactor at CEA-Saclay. In 2011 we plan to deploy the detector at the ILL research reactor in Grenoble. In 2012-13 we plan to deploy the detector at a power reactor, possibly under Safeguards regime, in collaboration with the IAEA.

# References

[1] "Final Report: Focused Workshop on Antineutrino Detection for safeguards Applications", report of IAEA Workshop, IAEA Headquarters, Vienna, Austria, Oct. 2008.

[2] "Saclay's neutrino detector concept for non proliferation safeguards applications", Lasserre, Th. September 24-26, 2006, Workshop., AAP 2006: Applied Antineutrino Physics.

[3] Reactor Neutrino Detection for Non Proliferation with the NUCIFER Experiment, A. Porta et al., IEEE proceedings 10.1109/ANIMMA.2009.5503653.

[4] The Nucifer experiment : reactor monitoring with antineutrinos for non-proliferation purpose, F. Yermia

et al. proceedings of GLOBAL 2009 International Conference, paper 9270, Paris, France.

[5] MURE : MCNP Utility for Reactor Evolution. O. MEPLAN et al., ENC Proceedings, Versailles, France, December 11-14 (2005).

[6] J.F. Briesmeister, MCNP - a General Monte Carlo N Particle Transport Code, Version 4C, LANL, LA-13709-M (2000).

[7] Improved Predictions of Reactor Antineutrino Spectra, T. Mueller et al., to be submitted to Phys. Rev. C[8] K. Schreckenbach et al., Determination of the antineutrino spectrum from 235U thermal neutron fission products up to 9.5 MeV, Phys. Lett. B160, 325-330, 1985;

Antineutrino Spectra from Pu-241 and Pu-239 thermal neutron fission products, A. A. Hahn et al., Phys. Lett. B218, 365-368, 1989.

[9] M. Fallot et al., these proceedings, contribution 246;

Nuclear Reactor Simulations for Unveiling Diversion Scenarios : capabilities of the antineutrino probe, M. Fallot et al., proceedings of GLOBAL 2009 International Conference, paper 9272, Paris, France.