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Technical Notes

Study on the large area MCP-PMT glass radioactivity reduction

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ABSTRACT

The Jiangmen Underground Neutrino Observatory (JUNO) will install approximately 18,000 20-inch Photomultiplier Tubes (PMTs) in the central detector. From the full detector Monte Carlo (MC) simulation, the PMT glass has one of the largest contribution to the natural radioactive background due to its large mass. Various techniques have been developed to reduce radioactivities introduced in the raw material and during the production process. Efforts such as low background raw material selection, dust and dirty water isolation and continuous production monitoring were taken to reduce the glass background. The radioactivity in the glass sample was measured using a low background gamma ray spectrometer equipped with a high-purity germanium (HPGe) detector. The average level of ^{238}U, ^{232}Th and ^{40}K in total MCP-PMT glass now reach 2.5 Bq/kg for ^{238}U, 0.5 Bq/kg for ^{232}Th and 0.5 Bq/kg for ^{40}K, at least a factor of 2.5 lower than other type of 20-inch PMTs.

1. Introduction

Natural radioactivity is one of the major backgrounds in various particle physics experiments with studies on neutrinos, dark matter or neutrinoless double beta decay, because of their rare interactions and low visible energies. Requirements on the selection of detector materials are always strict to achieve low background and extremely low energy threshold. In particular, Photomultiplier Tubes (PMTs) are widely used to detect the optical photons produced in the interaction between the incident particles and the detector, and their intrinsic radioactive background has to be controlled. For example, many studies on the PMT glass screening have been done in the dark matter experiments, such as LUX [1] and XENON [2]. In large-scale neutrino experiments, such as Super-Kamiokande [3], KamLAND [4], Borexino [5,6], Daya Bay [7] and JUNO [8], PMTs with diameters up to 50 cm (20 inch) are equipped and the development of such a large glass bulb with low radioactivity is extremely difficult due to the complex production technology.

The JUNO experiment aims to determine the neutrino mass hierarchy by detecting the oscillation of the antineutrinos from reactors. About 18,000 20-inch PMTs will be installed on the central detector to achieve $3\%/\sqrt{E(MeV)}$ energy resolution, and 2000 20-inch PMTs will be equipped in the water pool to record cosmic muons. A new type of 20-inch PMT with Micro-channel Plates (MCP) has been developed by the Institution of High Energy Physics (IHEP) and the North Night Vision Technology (NNVT) [9] in China. The JUNO detectors will use 5000 dynode-PMTs (Hamamatsu R12860-50) [10] and 15,000 MCP-PMTs. The radioactivity specifications of these two kinds of PMTs were found in the products data sheets and compared in Table 1.

2. The JUNO experiment

Reactor antineutrinos will be detected via the inverse beta decay process in the JUNO detector, with the signature of a prompt-delayed pair correlated in time, space and energy. About 60 antineutrinos per day [11] are expected after the event selection. The prompt energy is between 1 and 10 MeV, and the delayed energy is around 2.2 MeV, both of which have large overlap with the natural radioactivity spectra. Most of the signals from the radioactivity are *singles*, i.e., the isolated signals which have no physical correlation with others. However, when two singles accidentally fall into the correlated time and space windows, they can mimic the antineutrino events. According to the sensitivity study of mass hierarchy determination, the singles rate in the fiducial liquid scintillator target is required to be less than 10 Hz.

Various types of materials will be used in the central detector and the veto detector in JUNO. The full detector simulation of the main

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Table 1
Comparison of the radioactivity between two kinds of PMTs. The number of Hamamatsu PMTs is 4500 and of NNVT PMTs is 13,500, with the weight of each PMT assumed to be 7.4 kg. The radioactivity specifications were from the products data sheets, while their contribution to the singles rate in the central detector within the fiducial volume (FV) was scaled from Ref. [12]. Half of ^{40}K in the NNVT PMT is from the glass bulb while the rest is from the transition section glass and the stem glass, which will be discussed later.

Material	Mass	^{238}U	^{232}Th	^{40}K	Singles in FV(cps)
Hamamatsu	33 ton	4.96 Bq/kg	1.62 Bq/kg	10.8 Bq/kg	0.68
NNVT	100 ton	2.5 Bq/kg	0.5 Bq/kg	1.0 Bq/kg	0.79

materials has been done in Ref. [12] to obtain their contributions to the radioactive background. In the fiducial volume with the radius less than 17.2 m and below an energy threshold of 0.7 MeV, the singles rate contributed from Hamamatsu PMTs and NNVT PMTs were compared in Table 1. In the Monte Carlo simulation, a quarter of the PMTs in the central detector was Hamamatsu type and the rest was MCP-PMT type, while another 2000 PMTs in the veto detector were ignored in this table since they are far from the liquid scintillator volume. According to a further study on the mass hierarchy determination, the sensitivity in terms of $\Delta\chi^2$ can be increased by 0.1 with this PMT combination compared to the case that all PMTs have the same radioactivity as Hamamatsu's, because of less accidental backgrounds in the former case.

3. Radioactivity measurement system

In order to monitor the glass bulb production continuously, glass samples were taken directly from the furnace every day. These samples were delivered from the glass bulb factory to the IHEP to measure the radioactivity. Most of them can be measured in one week after their production.

3.1. High-purity germanium detector

The High-Purity Germanium (HPGe) detector is a high-precision γ spectrometer used to analyze the natural radioactivity in samples. Our spectrometer is a 4-inch well-type HPGe detector from Canberra [13]. The resolution is 2.1 keV at 1.33 MeV. The nominal size of the germanium crystal is 86.3 mm in diameter and 85.8 mm in height with a 4.89 mm front gap, and the thickness of the aluminum shell is 1.5 mm. Four layers of walls surrounding the germanium crystal were made for the γ background shielding and the anti-Compton measurement. From the inner layer to the outer layer, they are BGO crystal, oxygen-free copper, lead and plastic scintillator. Due to the limited size of the sample chamber in the HPGe spectrometer, the glass samples were made as cylinders with the dimensions of 120 mm in diameter and 50 mm in height.

3.2. Data analysis

The ^{238}U and ^{232}Th radioactivity are measured by their daughter nuclides in their decay chains, assuming the decay chains are at equilibrium. For ^{238}U, three daughter nuclides, known as ^{226}Ra (186.21 keV),^{214}Pb (295.22 keV, 351.93 keV) and ^{214}Bi (609.32 keV, 1120.29 keV), were measured. For ^{232}Th, another three daughter nuclides, known as ^{228}Ac (911.2 keV, 1459.14 keV),^{212} Pb (238.63 keV) and ^{208}Tl (583.19 keV, 2614.5 keV), were measured.

Four gamma sources (^{241}Am,^{133}Ba,^{57}Co,^{152}Eu) were used to calibrate the detector, with the energy ranging from 60 keV to 1460 keV. A Geant4 [14] simulation was used to correct the efficiencies of the HPGe detector system. The background spectrum was obtained by accumulating seven days of data without any test sample. The systematics uncertainty of the gamma spectrum measurement was estimated to be less than 0.5%, while the uncertainty of the efficiency correction caused by the irregular top surface of the measured sample (< 5 mm height fluctuation) was less than 5%. Nevertheless, the total uncertainty was limited by the statistics, which is ~15% in one day.

3.3. Radon decay in glass

The samples must be stored for at least 1000 h to reach equilibrium for the ^{238}U decay chain. The non-equilibrium is caused by the radon release due to its low solubility in the melted glass. As the melted glass gets cured, the radon releasing from the glass gets stopped and then the radioactivity chain returns to equilibrium. The recovery time depends on the remaining proportion of radon in the glass and the half life of ^{222}Rn. Assuming the remaining proportion of radon is m, a percentage from 0 to 100%, the radioactivities of ^{226}Ra and ^{222}Rn are expressed as

$$A_{^{226}Ra}(t) = \lambda_{Ra} N_0 e^{-\lambda_{Ra} t}, \tag{1}$$

$$A_{^{222}Rn}(t) = \frac{\lambda_{Ra} N_0}{\lambda_{Rn} - \lambda_{Ra}} (\lambda_{Rn} e^{-\lambda_{Ra} t} - (\lambda_{Rn} - m(\lambda_{Rn} - \lambda_{Ra})) e^{-\lambda_{Rn} t}), \tag{2}$$

where A_i is the radioactivity of i-nuclide, t is the time, λ_i is the decay constant of i-nuclide and N_0 is the initial number of atoms of ^{226}Ra. Given that $\lambda_{Rn} \gg \lambda_{Ra}$, the radioactivity ratio between ^{222}Rn and ^{226}Ra can be approximated as

$$Ratio = \frac{A_{^{222}Rn}(t)}{A_{^{226}Ra}(t)} \approx 1 - (1 - m) e^{-\lambda_{Rn} t}. \tag{3}$$

The ratio of radioactivity as a function of the time interval between the production and measurement of each sample is shown in Fig. 1. With the known half-life of ^{222}Rn, λ_{Rn} can be either fixed at the true value or left as a free parameter to fit, then the remaining proportion of radon m is fitted. The two fitting curves are consistent and the best fit values of m have only 1σ difference. The time internal between the production and the measurement of each glass sample was recorded and used to judge if the sample is at equilibrium or not. When the glass was at equilibrium, the average of ^{214}Pb and ^{214}Bi measurements was used due to its higher precision. Otherwise, the result of ^{226}Ra was used to remove the non-equilibrium effect.

4. Radioactivity control to PMT glass

The schematic diagram of a typical MCP-PMT, as well as the key dimensions, are shown in Fig. 2. Three components of the NNVT PMT use glass, including the bulb, the transition section and the stem. The 20-inch glass bulb is made from a glass furnace, and its fabrication is a kind of handicraft. The glass bulb is sealed with kovar metal on the bottom, which has a very different thermal expansion coefficient from the glass. To avoid damages to the glass-kovar connection, a transition section has been designed and it consists of four types of glass with different thermal expansion coefficients. The coefficients are affected by the covalent bond of the glass and can be adjusted by different doping of ions.

The main components of a typical MCP-PMT are shown in Table 2. The total mass fraction of the glass compositions is greater than 92%. Thus the radioactivity control to each glass component is critical to reduce the overall radioactivity of the PMT. After extensive tests of the raw materials and the radioactivity control approaches, as well as Monte Carlo simulations, we have set the radioactivity specifications to each component of the PMT, also as shown in Table 2. The radioactivity control to each glass component of the PMT is discussed below.