Radon/Radium measurement from Gd-loaded water

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Abstract.

The Super Kamiokande-Gd project (SK-Gd) aims to load Gd powder into the SK water in order to detect the neutrons produced by $\nu$ and $\bar{\nu}$ interactions in the water. Despite Gd powder's high purification, some radio-isotope contaminates are expected. Among them $^{222}\text{Rn}$, from $^{226}\text{Ra}$ decays, is a threat for high-precision low energy physics experiments like the Super-Kamiokande experiment (SK). The measurement of $^{222}\text{Rn}$ from Gd-water is then needed in order to allow monitoring of its concentration in the water, as well as to test the efficiency of radio-isotope removal methods still in development. Here we present the system used to determine the $^{222}\text{Rn}$ concentration from pure water and Gd-water. We also show the measurements of $^{222}\text{Rn}$ ($^{226}\text{Ra}$) concentration from water loaded with different Gd powders.

1. Introduction

Super-Kamiokande (SK) is a successful neutrino experiment located in the Kamioka mine, about 1 km below the top of Ikenoyama, in Kamioka-cho (Japan). SK is a water Cerenkov detector, filled with 50 kt tons of pure water, operating since 1996. The SK collaboration is now planning to load gadolinium (Gd) in the water of the detector in order to greatly improve the detectability of the neutrons produced by some (anti)neutrino interactions. The main goal of the SK-Gd project is the detection of the Supernova Relic Neutrino signal (SRN) using the inverse beta decay reaction ($\bar{\nu}_e + p \rightarrow e^+ + n$), which produces a neutron together with a positron.

In low energy physics, like the solar neutrino analysis or the SRN analysis in SK, the radiopurity of the detector component is a critical issue. In SK the main issue is the radiopurity of the water, hence we need to continuously remove the radioisotopes dissolved in the water from the environment like uranium or radium. In order to remove these radioisotopes from the water, cation and anion exchange resins are usually used. In the case of the SK-Gd project, the Gd is loaded as Gd sulfate, therefore, the standard cation exchange resin would also remove the Gd. A special cation exchange resin has been developed in order to remove cations (like radium) without removing Gd.

In order to test the capacity of this resin, we developed a system able to measure the concentration of radium in the water from pure-water or Gd-loaded water. This system consists of a Gd-water production and purification system, able to process 1 m$^3$ of water, and of a radon measurement system. The system takes advantage of the fact that secular equilibrium between $^{226}\text{Ra}$ and $^{222}\text{Rn}$ (thereafter noted “radon”) can be achieved within a few weeks.
2. Gd-water preparation and Radium measurement

The system we are using to produce Gd-loaded water and to measure its radium concentration is based on the Super-Kamiokande water and radon measurement systems, described in [1].

The water production and purification system is shown in Figure 1. It consists of a small version of the SK water purification system, using filters, a membrane degasifier and a UV lamp. Placeholders for the anion and cation resins are already installed but have so far been bypassed. Gd-loaded water is produced in the Gd mixer, by mixing Gd-powder and pure-water. 1 ton of water can be processed by the system. The water is circulated for 24h in the purification system, allowing to remove most of the Rn in the water. The water then rests in the 1 m$^3$ tank for at least 20 days, in order to reach secular equilibrium between $^{226}$Ra and $^{222}$Rn.

Figure 1. Scheme of the Gd-water production and purification system

The radon measurement system is shown in Figure 2(A), it is based on the system used to monitor the Rn concentration in the water of the SK detector [1]. It consists of a radon mixer tower, in which the water from the 1 m$^3$ tank is injected. Its structure is such that the radon is expected to degasify from the water with an efficiency of 65 %. The radon is then extracted outside the mixer via a pure air (G1 class) flow, passing through cooled copper, in order to reduce the humidity, and through a cooled activated charcoal, in which the radon is expected to be trapped with 99 % efficiency. After three hours of such processing, aiming to increase the radon concentration in the charcoal, the water circulation is stopped, and we start the radon extraction from the activated charcoal. As shown on Figure 2(B), the charcoal is heated to 180 °C, allowing extraction of the radon and collection in a high precision radon detector [2].

Figure 2. Scheme of the radon measurement system. Radon is noted “Rn”, Copper, “Cu”, and Charcoal, “C”. A) Radon concentration phase. B) Radon extraction phase.

3. Results and discussion

Table 1 shows the measured radon concentration for the different samples we used. We measured pure-water samples, as well as Gd-loaded water samples. The pure-water samples (samples #1
and #2) were used to determine the background (BG) level of the measurement system. Using “clean” Gd-powders (Gd-A and Gd-B used in samples #3 and #4), like those to be loaded in Super-Kamiokande, we found out that the radium concentration is too low to allow us to measure a significant excess compared to the BG level of the system. We therefore tested our system with radium-loaded Gd-powder (sample #5) and “dirtier” Gd-powder (Gd-C in samples #6 and #7). Using these powders with high radium concentrations, a significant excess with respect to the BG level can be observed. This result will allow us to test the effect of the future special cation exchange resin with such high radium concentration powder.

| Samples         | $\rho_{\text{Water meas.}}$ (mBq/m$^3$) | $\rho_{\text{Powder meas.}}$ (mBq/kg) | $\rho_{\text{Powder Expected}}$ (mBq/kg) |
|-----------------|----------------------------------------|---------------------------------------|------------------------------------------|
| #1 Pure-water (BG) | 1.9 ± 0.3                             | n/a                                   | n/a                                      |
| #2 Pure-water (BG) | 1.3 ± 0.3                             | n/a                                   | n/a                                      |
| #3 Gd-A (clean)   | 0.4 ± 0.6                             | 0.2 ± 0.3                             | ∼ 0.2                                   |
| #4 Gd-B (clean)   | 0.2 ± 0.6                             | 0.1 ± 0.3                             | ∼ 0.2                                   |
| #5 Gd-A (Ra-loaded) | 3.4 ± 0.7                             | 1.6 ± 0.4                             | ∼ 6.7                                   |
| #6 Gd-C (w/ Air)  | 8.9 ± 0.9                             | 3.6 ± 0.4                             | ∼ 12                                    |
| #7 Gd-C (w/o Air) | 13.7 ± 1.2                            | 5.5 ± 0.5                             | ∼ 12                                    |

Table 1. Measured radon concentrations in water ($\rho_{\text{Water meas.}}$), estimated radium concentrations in the Gd-powder ($\rho_{\text{Powder meas.}}$), and expected radium concentrations in the Gd-powder ($\rho_{\text{Powder Expected}}$).

Contrary to expectations, when we compare the expected radium concentration in the powder, obtained with germanium detectors, and the estimation from the radon measurement in the water, we found a clear deficit. The estimation from the radon measurement in the water gives $\sim 2 - 3$ time lower radium concentration with respect to the measurement from a germanium detector. This issue is not yet fully understood. Currently, the most probable explanation is a degasification of the radon from the water inside the 1 m$^3$ tank: The literature [3] indicates the existence of fast radon exchange between water and air, which could explain our observations. Degasification should happen during the rest period and during the measurement itself.

We performed several measurements in order to test the impact of a degasification occurring during the rest period by activating or stopping the air circulation in the 1 m$^3$ tank (samples #6 and #7). Our results indicated a small increase when there is no air circulation. However, this increase is not large enough to explain the discrepancy from the expectation. We are currently trying to determine what would be the impact of a radon degasification in the 1 m$^3$ occurring during the measurement itself if fast radon exchange between water and air exist.

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