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Preliminary study for decay time analysis of TCSPC and radiation measurement system

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1. Introduction

The radiation measurement system is vital for the actual issue in nuclear facility accident preparation, decommissioning nuclear facilities, regular environmental radiation monitoring, medical, nuclear security and safeguards. In particular, a large-volume measurement system is required in the field of nuclear facilities decommissioning and environmental radiation monitoring. When dismantling a nuclear facility, radiological characterization and radiation monitoring must be performed. After decommissioning, it must be proven that the residual contamination level of the decommissioning site is below the release criteria. For this purpose, plastic scintillator with good process-ability and a short decay time is widely used, but the plastic scintillator has poor resolution, so radionuclide analysis is not possible. Currently, many studies have been conducted on large-volume plastic detection systems [1,2]. Time-resolved photon counting is an essential analysis method in the field of precision measurement such as classical and quantum domains. Among them, time-coupled single-photon counting (TCSPC) is a key technology for applications such as fluorescence life spectroscopy and microscope, time-open/close Raman spectroscopy, time-off light 3D imaging, light-in-fly imaging, and computer diffusion optical tomography. By continuously accumulating the detected signal on the time axis, an attenuation curve can be created. When the intensity of fluorescence is high, many photons are detected, and when the intensity of fluorescence is weak, fewer photons are detected. By plotting this histogram, a graph similar to the original attenuation curve can be obtained called TSCPC. Such photon counting equipment requires a fast pulse laser with sharp pulse width of picoseconds or femtoseconds as a light source. Microchannel plate (MCP)-photomultiplier tubes (PMT) are mainly used as detectors. If multiple photons arrive at the detector at once, the detector records only the first time it arrived. Therefore, the intensity of the light source must be adjusted so that no more than one photon is generated per pulse. In general, the amount of light is adjusted so that one photon is detected per 100 pulses [3,4]. In this study, Nd:YAG Laser TCSPC equipment was built to analyze the fluorescence characteristics of scintillators with various sizes and shapes, and an optical sensor scintillator containing cadmium-doped zinc oxide (CZO) nanomaterial was manufactured and demonstrated with Nd:YAG laser TCSPC equipment. Finally, it was demonstrated a process, which means an integrated apparatus for decay time analyzer and gamma radiation measurement with a liquid scintillator homemade based CZO nanomaterial that could trigger a Compton edge energy.

2. Methods and Results

2.1 Fabrication of a liquid scintillator

The liquid scintillator used in this study was placed in a 35 mL microcuvette and the timing properties were analyzed. The microcuvette has an inside diameter of 47 mm and an inside thickness of 20 mm. It was manufactured by adding PPO and POPOP to toluene. CZO nanomaterial was added to improve an efficiency by increasing the reaction to photons. First, the sample was stirred for 5 hours by adding toluene (>98 %), PPO (0.4 wt%), POPOP (0.01 wt%), and CZO (0.1 wt%). The microbubbles generated through the stirring process were removed through the degassing process for 5 hours. In addition, the liquid sample was carefully placed in a 35 mL microcuvette and sealed. A CZO-loaded liquid scintillator and a basal liquid scintillator without CZO were fabricated. Since CZO was employed at a high concentration (100 mg/mL) from the supplier, it was diluted 1/100 with toluene and used. Absorbance and transmittance were measured three times by randomly selecting ten points of a 50 m diameter samples and then averaged. When light passes through, the intensity of the light decreases because the light is absorbed by the sample. The amount of light that has passed through the sample (Transmittance, T) is expressed as the ratio of the intensity of light (I0) when there is no light absorbing material and the intensity (I) of light when there is a light absorbing material. It is called transmittance and is expressed as $T=I/I_0$. The absorption rate of the CZO-loaded liquid scintillator is larger at wavelength above 400 nm. CZO-loaded liquid scintillator (toluene+PPO+POPOP+CZO) transmits over 400 nm like the basal liquid scintillator (toluene+PPO+POPOP), transmittance was lower than that of the basal liquid scintillator due to the effect of quantum dot. Photo-luminescence was measured using a Spectrophotofluorometer and analyzed at excitation wavelengths of 264 nm and 316 nm, respectively. It shows emission wavelength at about 420 nm. And quantum dots shows an emission wavelength at about 450 nm. There show weak peaks around 440 nm, which appears to be the effect of the secondary nanomaterial (POPOP), and the effect was not observed.

2.2 Set-up of TCSPC system and radiation measurement system

The TCSPC system and radiation measurement system were constructed simultaneously. Figure 1 shows a schematic diagram of the manufactured TCSPC system and radiation measurement system. Decay time analysis system to observe the optical properties of a liquid scintillator to which CZO nanomaterial are demonstrated, and this system can analyze samples of various sizes, shapes and types without additional sampling. TCSPC system was constructed using Laser(3rd Harmonics of Q-switched Nd:YAG laser, Brilliant B), and the main schematic diagram of the constructed TCSPC system is shown in Figure 1 (left). This system is equipped with a band filter and a dichroic mirror to control the excitation wavelength of light. The Dichroic mirror separated Nd:YAG light into 1064 nm, 532 nm, and 355 nm. Light with wavelength of 355 nm passed through the sample, and the signal detected by the PMT(R928, HAMAMATSU) was transmitted to the oscilloscope(DS6104, RIGOL) to analyze the output data from the oscilloscope. The oscilloscope stores one sample point per waveform to reduce noise. The average value of 64 collected data was used, and 10ns pulse data were obtained. All experiments were performed in a dark box to block unnecessary light. Not only the decay time analysis but also the radiation detection system was constructed so that the decay time and scintillation counting of the liquid sample could be measured quickly.

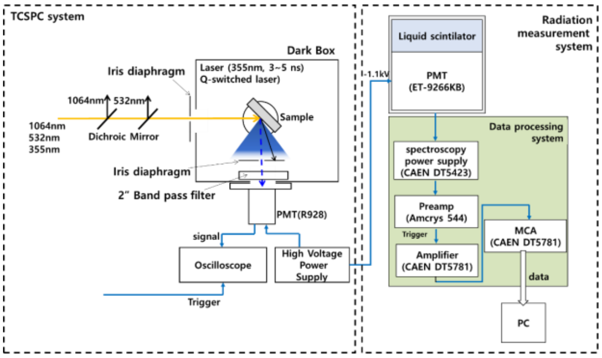

Figure 1. A schematic diagram for decay time analysis of TCSPC and radiation measurement system

Cs-137 point source was used, and data were analyzed based on Compton edge due to inherent resolution of plastic scintillator. The radiation source was located at 20 mm from the liquid sample. Liquid scintillator was connected with 2 inch PMT (ET-9266KB, ET-Enterprises Ltd.). Signals extracted from PMT were processed by Preamp (Amcrys 544, Amcrys), Amp (DT5781, CAEN) and MCA (DT5781, CAEN) shown in Figure 1 (right). TCSPC system and radiation measurement system were directly constructed for the fast analysis of samples of various sizes, shapes, and types. The homemade TCSPC system and radiation measurement system are shown in Figure 2. Figure 2(a) shows a TCSPC system and Figure 2(b) shows a radiation measurement system. In Figure 2(a), all the components used are marked, and the sample part blocks unnecessary light by using a Dark Box. Nd:YAG Laser was used as the light source of the homemade TCSPC system. The laser light divided by the dichroic mirror excites the sample, and the pho-ton emitted from the excited sample is detected by the PMT and converted into an electrical pulse. In addition, electrical pulses were analyzed through an oscilloscope. In Figure 2(b), radiation measurements system is located next to the TCSPC system and is configured so that it can be measured immediately from an integrated apparatus.

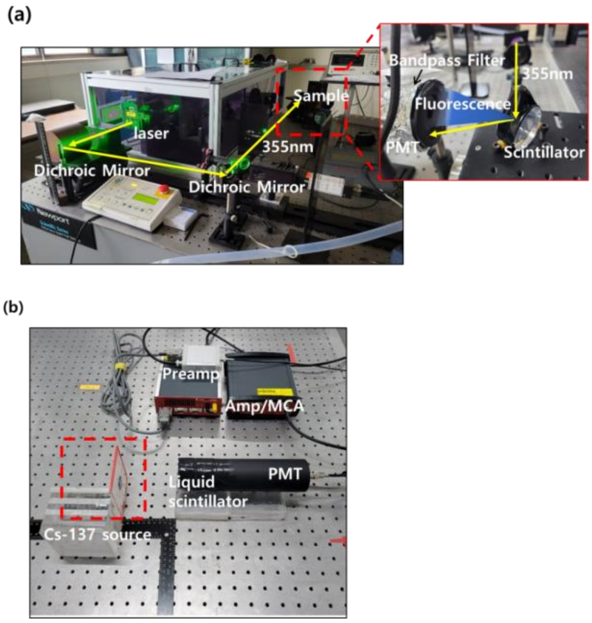

Figure 2. Setup of decay time analysis system using the Nd:YAG laser(a) and radiation measurement system (b)

3. Conclusions

Time resolution can be obtained in various methods depending on the required sensitivity and time domain. Time domain methods are more diverse than frequency domain methods for measuring the lifetime of fluorescence. A representative method is time-correlated single photon counting. It measures the time the photons arrive after excitation which means to obtain a decay curve by accumulating the data on the time axis. A homemade TCSPC system capable of analyzing decay time of various samples was constructed. This system used Nd:YAG Laser, dichroic mirror, and band filter to irradiate the sample with appropriate light. Then, the signal was analyzed using a PMT and an oscilloscope. Unlike commercial TCSPC equipment, homemade TCSPC system can analyze samples of various sizes, shapes, and types. In addition, to evaluate the reliability of this system, Base and CZO loaded liquid scintillator were fabricated and decay time was measured. When comparing the results with commercial TCSPC equipment, it was confirmed that the error was within 5%. Therefore, the applicability of homemade TCSPC system was demonstrated for the purpose of analyzing the timing performance in nanoseconds. In the future, if the PMT of this system is configured as Fast PMT, it is expected that the timing performance in picoseconds is also possible to analyze.

As a result of performing a measurement experiment using the Cs-137 source, it was analyzed that the liquid scintillator containing CZO was shifted to lower energy than the liquid scintillator without the CZO representing the Compton edge energy of Cs-137 is 477.3 keV, which hardly generates photoelectric effect, and mainly Compton scattering occurs. Finally, a system for radiation measurement and decay time analysis was directly constructed using a manufactured liquid scintillator homemade based on basic toluene and nanomaterial. Moreover, this process on a system can directly configure and measure the properties of liquid scintillators of various sizes and different shapes from an apparatus for decay time analyzer and radiation measurements.

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