Development of Fast Scintillation Materials for High-Energy X-Ray Detection

We successfully developed fast scintillation materials for high-energy X-ray detection using two approaches. One is based on plastic scintillators; the detection efficiency of plastic scintillators for high-energy X-ray photons was significantly enhanced by loading Hf-based oxide nanoparticles. The other is the use of Auger-free luminescence in halide crystals. Among those, we found that Cs_2ZnCl_4 exhibits relatively efficient Auger-free luminescence and a negligible long scintillation component. These materials can be used for high-energy X-ray detection in the case that detection signals having fast decay and no long tail (afterglow) are desired.

There is an increasing demand for fast detectors for high-energy X-ray detection. For X-rays whose energy is on the order of several tens of keV, scintillation detectors equipped with fast scintillators would meet such demand. Among several approaches to develop such fast scintillation materials, we introduce the development of scintillation materials based on two approaches.

One is based on plastic scintillators; plastic scintillators are known to exhibit fast scintillation whose decay time constant is on the order of nanoseconds. However, a disadvantage of plastic scintillators is low atomic numbers of constituent elements, leading to a low interaction probability with X-rays or gamma-rays. The low interaction probabilities result in low detection efficiency. Loading of high atomic number elements is an effective method to enhance the average atomic number and thus the detection efficiency for high-energy photons. The conventional method is to load organometallic molecules containing heavy elements. However, such molecules often act as quenching centers, and loading a small amount of such molecules severely degrades the light yield. We have developed an alternative approach to enhance the average atomic number: loading of metal oxide nanoparticles [1, 2]. Here, we introduce the development of plastic scintillators loaded with Hf-based oxide nanoparticles [2]. Figure 1(a) shows the TEM image of plastic scintillator loaded with Hf-based oxide nanoparticles at 10 wt%. The particle size was less than 300 nm. The size of the nanoparticles was smaller than the wavelength of scintillation photons of ~400 nm. Thus, the scintillator was transparent to the scintillation photons. Figure 2(b) shows the temporal profile of scintillation of the plastic scintillator loaded with Hf-based oxide nanoparticles. The decay time constant of the scintillation was 2.5 ns, which is comparable to that of generic plastic scintillators. In addition, long scintillation components were negligible. The detection efficiency for 67.4 keV X-ray photons was twice that of a commercial plastic scintillator for high-energy photon detection, NE-142 (loaded with Pb at 5 wt%). Furthermore, the light yield was 1.2 times higher than that of NE-142. Based on these results, we successfully enhanced the detection efficiency of Hf-based oxide nanoparticles without degrading the light yield and excellent timing property.

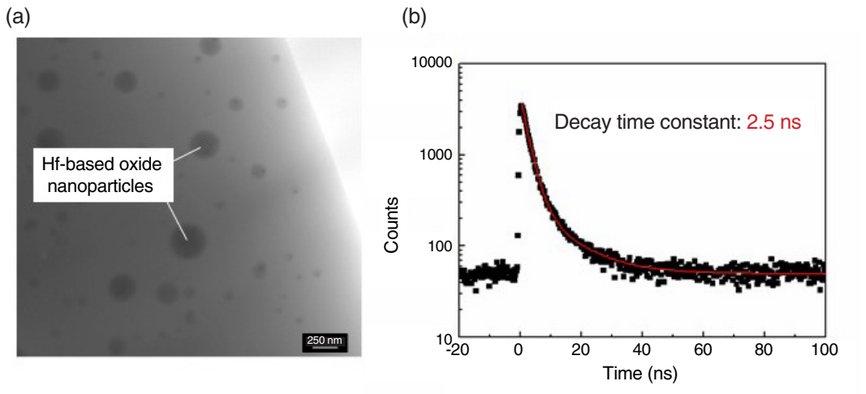

Figure 1: (a) TEM image of plastic scintillator loaded with Hf-based oxide nanoparticles. (b) Temporal profile of scintillation of the plastic scintillator loaded with Hf-based oxide nanoparticles.

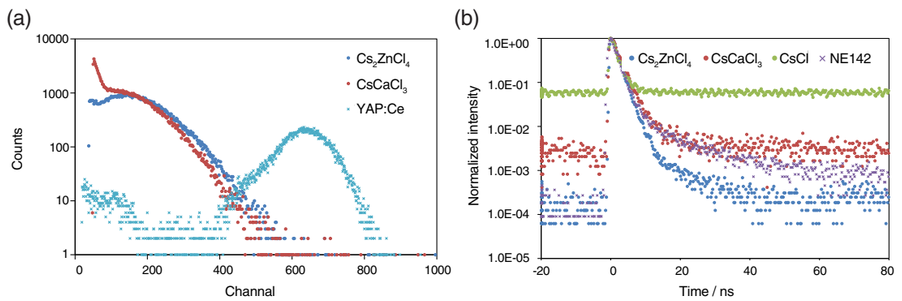

Figure 2: (a) Pulse height spectra of Cs_2ZnCl_4, $CsCaCl_3$, and YAP:Ce. The amplifier gain for Cs_2ZnCl_4 and $CsCaCl_3$ was five times that of YAP:Ce. (b) Temporal profiles of Cs_2ZnCl_4 and other similar compounds. The temporal profile of NE-142 is also shown.

The other approach is based on the Auger-free luminescence, which is a radiative process between the valence band and the outermost core level. In most insulators, holes at the outermost core level decay via the Auger process, in which the recombination energy between the core hole and a valence electron is deposited to another valence electron, i.e., an Auger electron. In contrast, in some insulators, the Auger process is prohibited owing to the band structure, and the recombination energy is released via photon emission, and this photon emission process is the Auger-free luminescence. The decay time constant of the Auger-free luminescence is subnanosecond to nanosecond, making it suitable for fast scintillation. A well-known scintillator exhibiting the Auger-free luminescence is BaF_2. However, BaF_2 has a disadvantage that it also exhibits a slow scintillation component with a decay time constant of 600 ns. We searched for insulators exhibiting the Auger-free luminescence without such long component, and developed several scintillators exhibiting Auger-free luminescence [3, 4]. Among them, we found that Cs_2ZnCl_4 exhibits relatively efficient scintillation due to the Auger-free luminescence and negligible long components [4]. Figure 2(a) shows the pulse height spectra of Cs_2ZnCl_4, $CsCaCl_3$, and YAP:Ce. The amplifier gain for Cs_2ZnCl_4 and $CsCaCl_3$ was five times that of YAP:Ce. Based on the peak positions and the wavelength-dependent sensitivity of the photomultiplier tube, the light yield of Cs_2ZnCl_4 was estimated to be 630 photons/MeV. The light yield of $CsCaCl_3$ is estimated to be quite similar to that of Cs_2ZnCl_4. Figure 2(b) shows the temporal profiles of Cs_2ZnCl_4 and other similar compounds. The temporal profile of NE-142 is also shown. The contribution of components having a decay time constant longer than the period of successive pulses in the single bunch oparation mode of the Photon factory (624 ns) can be seen as the time-independent component. It is clearly seen that Cs_2ZnCl_4 had a much lower contribution of long component than those of other scintillators. The main component of Cs_2ZnCl_4 had a decay time constant of 1.9 ns.

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BEAMLINES

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