

# Scintillation Properties of $Ce^{3+}$ Doped Silicon-Magnesium-Aluminum-Lithium Glass Scintillators by using Radiation Sources

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Glass scintillators can be doped with different elements to improve their luminescence properties. In this study, we present 0.5 wt%  $Ce^{3+}$ -doped  $(58-x)SiO_2-4MgO-18Al_2O_3-20Li_2O$  and  $(58-x)SiO_2-4MgO-18Al_2O_3-20LiF$  glass scintillators. We used a pulsed laser to measure the laser-induced emission spectrum and the decay time with decreasing temperature from 300 K to 10 K. The light intensity of both glass samples increased with decreasing temperature from 300 K to 10 K. Additionally, the two glass samples exhibited a fast decay time of approximately 25 ns. In the photo-induced spectrum, an excitation peak at 312 nm and two emission peaks, at 370 nm and 700 nm, were observed in both the glass samples. The X-ray-induced emission intensity of former is approximately six times higher than that of latter, and only former glass sample exhibits a proton-induced emission spectrum.

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## I. INTRODUCTION

Glass scintillators can be easily fabricated in large sizes and various shapes [1]. They can also be fabricated with various doping materials by using a melt-quenching method [2]. Specifically,  $Ce^{3+}$  is a rare-earth material with a  $4f-5d$  transition. Therefore, glass scintillators doped with  $Ce^{3+}$  have a fast decay time corresponding to a few nanoseconds and an emission wavelength in the visible range of 300 nm to 600 nm [3]. These properties potentially indicate that glass scintillators doped with  $Ce^{3+}$  can be used in particle physics, nuclear physics, astrophysics, and medical imaging [4].



Fig. 1. (Color online) Glass samples :  $(58-x)SiO_2:4MgO:18Al_2O_3:20Li_2O:xCeF_3$  (left) and  $(58-x)SiO_2:4MgO:18Al_2O_3:20LiF:xCeF_3$  (right).

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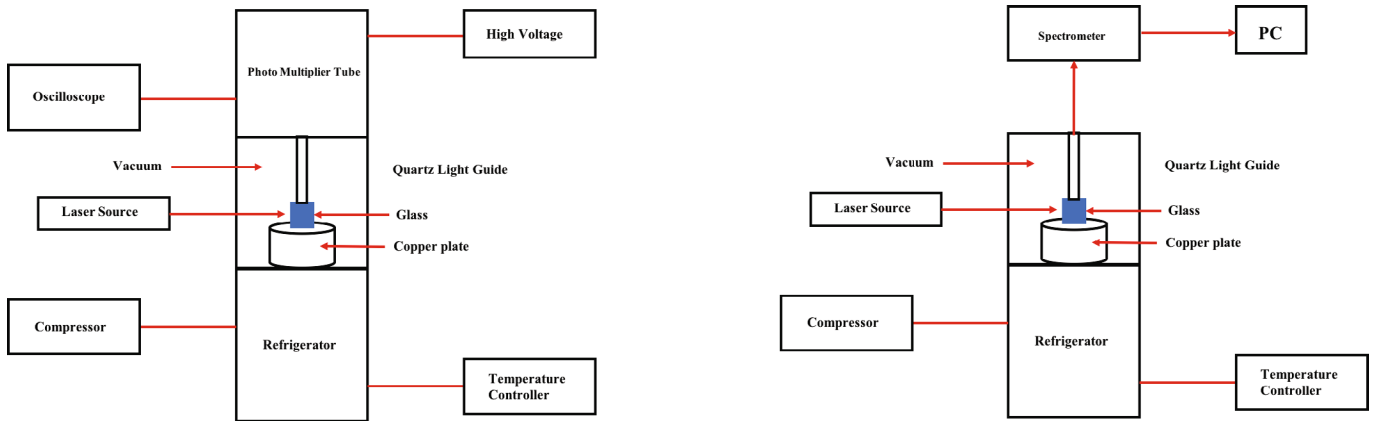


Fig. 2. (Color online) Schematic of the experiment setup for measuring the decay-time (left) and the laser-induced emission spectrum (right) with changing temperature.

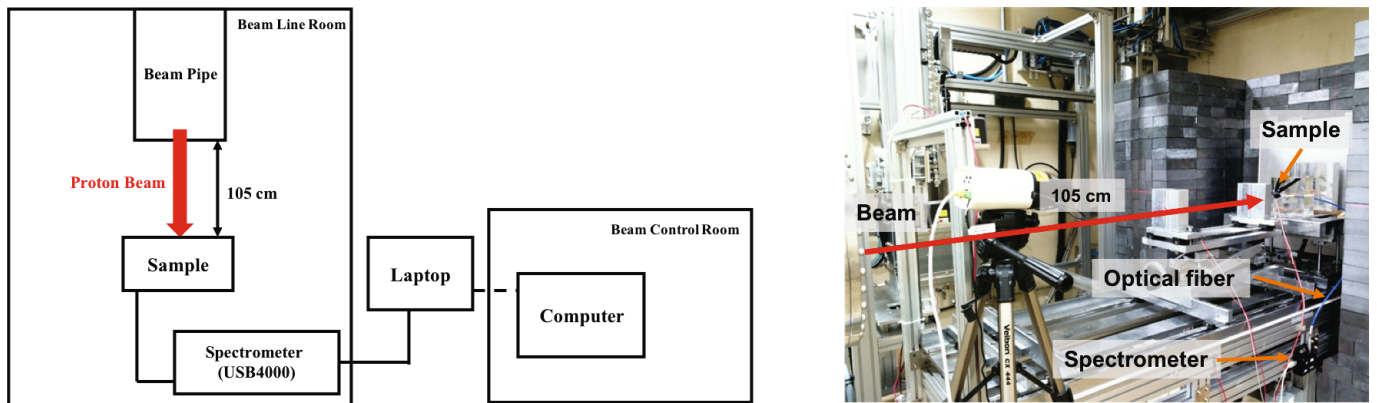


Fig. 3. (Color online) Schematic of the experimental setup for the 100 MeV proton-induced emission spectrum (left) and photograph of beam line room (right).

Our glass samples had differences in the host materials. One of the glass samples had Li<sub>2</sub>O while the other glass sample had LiF in the host material. Both glass samples included Li in the host material; thus, they can be used for neutron detection [5]. Although they had equal weight percents of Ce<sup>3+</sup>, they exhibited different luminescence properties because they were composed of different host materials [6]. Their luminescence properties, such as intrinsic defect luminescence, were affected by the dopant materials and the host materials [7]. We used UV laser light, light, X-rays, and protons to study the luminescence properties of glass scintillators. Additionally, we studied the luminescence properties of a glass scintillator at low temperatures.

## II. FABRICATION AND MEASUREMENT SYSTEM

Glass samples with chemical compositions of (58-*x*)SiO<sub>2</sub>:4MgO:18Al<sub>2</sub>O<sub>3</sub>:10Li<sub>2</sub>O:*x*CeF<sub>3</sub> (SiMgAlLi<sub>2</sub>O:Ce glass) and

(58-*x*)SiO<sub>2</sub>:4MgO:18Al<sub>2</sub>O<sub>3</sub>:20LiF:*x*CeF<sub>3</sub> (SiMgAlLiF:Ce glass), in which *x* is 0.5 wt%, were prepared by using a melt-quenching technique at the Center of Excellence in Glass Technology and Materials Science in Thailand. In this process, 30 g of a homogeneous mixture of starting chemicals, such as SiO<sub>2</sub>, MgO, Al<sub>2</sub>O<sub>3</sub>, Li<sub>2</sub>O, and LiF, are placed in high-purity alumina crucibles and melted at 1400 °C for 3 h by using an electrical furnace. These melted materials are quenched in pre-heated stainless steel molds. This is followed by annealing at 550 °C for 3 h to remove the thermal strain. Finally, the SiMgAlLi<sub>2</sub>O:Ce and SiMgAlLiF:Ce are cut and polished to samples with dimensions 9.9 mm × 10.6 mm × 5.2 mm and 9.6 mm × 10.2 mm × 5.1 mm, respectively [8]. Figure 1 shows each of the glass samples.

A laser with a pulse width of 10 ns and a repetition rate of 6.06 kHz (CNI, 266 nm UV solid state laser, MPLF-266) was used to measure the laser-induced emission spectrum and the decay time. Figure 2 (left) shows the experimental setup for measuring the decay time at low temperature. The glass sample is located on the copper plate. The quartz light guide is attached to the top face of the glass sample. The laser source irradiates to the

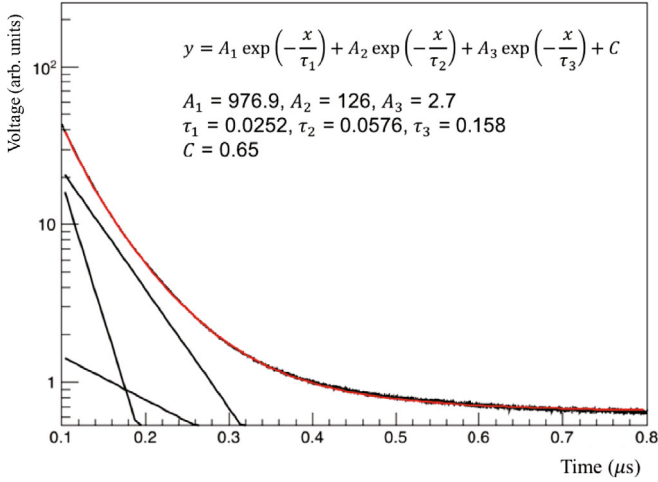


Fig. 4. (Color online) Decay time for SiMgAlLiF:Ce at 100 K. A pulsed laser was used to measure the decay time.

surface of the glass sample. The light emitted from the glass sample is transmitted through a quartz guide to a photomultiplier tube (Hamamatsu, H6610) and oscilloscope (Lecroy, 610Zi). The temperature ranges from 300 K to 10 K. Figure 2 (right) shows the experimental setup for obtaining the laser-induced emission spectrum. The quartz light guide is coupled with the optical fiber. The light emitted from the glass sample is measured by using a spectrometer (QE65000, Ocean Optics Co.).

### III. RESULTS AND DISCUSSION

#### 1. Decay time

The X-ray-induced emission spectra of the glass samples were measured at room temperature. In order to measure the emission spectrum, we used X-rays from an X-ray tube with a W anode (DRGEM Co.). The power settings of the X-ray tube are set to 100 kV and 2.0 mA. The X-ray-induced emission spectra of the glass samples are measured by using the QE65000 spectrometer. The proton-induced emission spectrum is measured by using the proton beam at the Korea Multi-purpose Accelerator Complex (KOMAC) [9]. Figure 3 shows a schematic diagram of the experimental setup for obtaining the proton-induced emission spectrum and photograph of the proton beam line with glass sample. The energy of the proton beam is 100 MeV, and the absorbed dose is about 8 Gy in each glass sample. The distance between the beam pipe and the sample is 1.5 m. The glass sample is attached to an optical fiber by using teflon tape and aluminum foil. The other side of the optical fiber is attached to the optical spectrometer (USB4000, Ocean Optics Co.). Excitation and emission spectra are measured by using a xenon lamp (HORIBA Jobin Yvon, FL3-2iHR). The

Table 1. Decay times of the SiMgAlLi<sub>2</sub>O:Ce and the SiMgAlLiF:Ce samples at different temperatures.

Temperature (K)	SiMgAlLi <sub>2</sub> O:Ce			SiMgAlLi <sub>2</sub> F:Ce		
	$\tau_1$ (ns)	$\tau_2$ (ns)	$\tau_3$ (ns)	$\tau_1$ (ns)	$\tau_2$ (ns)	$\tau_3$ (ns)
300	24.8	64.0	279	23.7	57.4	199
250	28.3	66.0	202	23.8	57.0	187
200	25.5	69.0	298	23.2	51.9	109
150	27.1	66.7	211	23.9	55.9	153
100	24.7	66.9	240	25.2	57.6	158
50	25.2	69.6	264	25.3	55.3	134
10	25.2	69.7	263	25.7	60.3	277

xenon source is placed at a right angle to the photo detector; thus, the glass samples are placed at 45° relative to the incident direction of the xenon source.

A pulsed laser source, refrigerator, and oscilloscope were used to measure the decay time of the glass samples at a low temperature. The decay time is measured by changing the temperature from 300 K to 10 K. Figure 4 shows three decay components in the SiMgAlLi<sub>2</sub>O:Ce glass. We fit the decay time curves by using three exponential functions defined as follows:

$$y = A_1 \exp(-x/\tau_1) + A_2 \exp(-x/\tau_2) + A_3 \exp(-x/\tau_3) + C, \quad (1)$$

where  $A_1$ ,  $A_2$ , and  $A_3$  denote the intensity contributions and  $\tau_1$ ,  $\tau_2$ , and  $\tau_3$  denote these decay time constant. Table 1 shows each decay time component of SiMgAl<sub>2</sub>O:Ce and SiMgAlF:Ce at various temperatures. No significant difference in the decay time with changing temperature was observed in these two glass samples. The two glass samples exhibited similar high decay times given that the glass samples had same weight percents of Ce<sup>3+</sup>.

#### 2. Photo-induced excitation and emission spectra

In order to examine the luminescence properties of SiMgAlLiF:Ce and SiMgAlLi<sub>2</sub>O:Ce, we measured the photo-induced excitation and emission spectra. Figure 5 shows the excitation and emission spectra of the glass samples. The black solid and blue dotted lines indicate the excitation spectrum, and the red dotted line indicates the emission spectrum of the glass samples. Two emission peaks and an excitation peak are observed in both glass samples. SiMgAlLiF:Ce exhibits an excitation peak at 312 nm and two emission peaks, are each at 360 nm and 680 nm. The 4*f*-5*d* transition indicates an excitation peak at 312 nm and an emission peak at 360 nm. The emission peak at 680 nm is caused by the defect luminescence of the SiMgAlLiF:Ce glass [7]. In

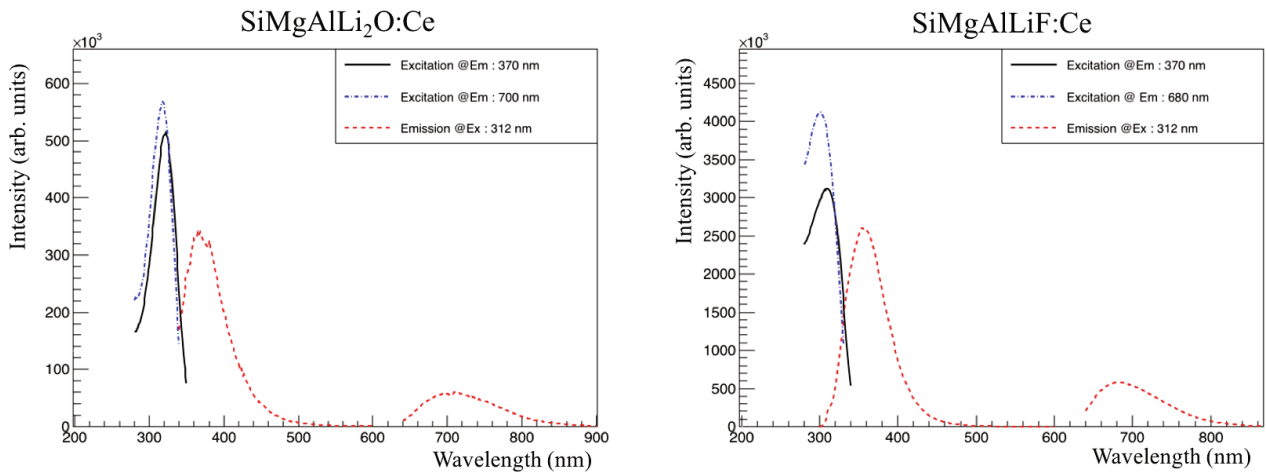


Fig. 5. (Color online) Photo-induced excitation and emission spectra of SiMgAlLi<sub>2</sub>O:Ce (left) and SiMgAlLiF:Ce (right). The black solid line denotes the excitation spectrum, and red dotted line denotes the emission spectrum. An excitation peak and two emission peaks are observed in each glass sample.

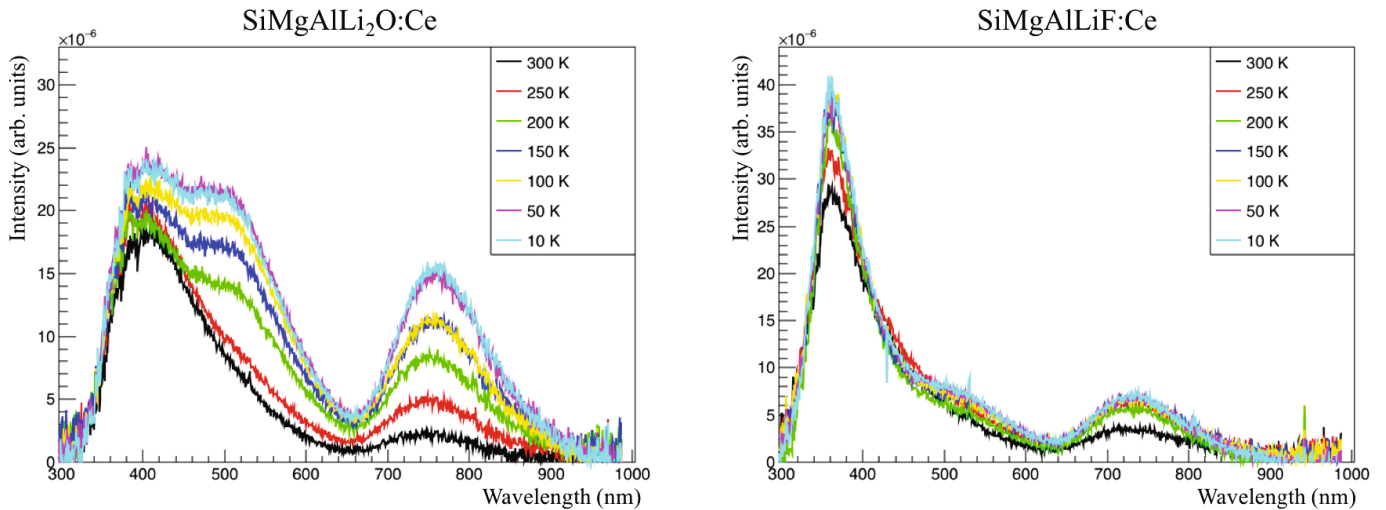


Fig. 6. (Color online) Laser-induced emission spectrum of SiMgAlLi<sub>2</sub>O:Ce (left) and SiMgAlLiF:Ce (right) for various values of the temperature.

comparison, SiMgAlLi<sub>2</sub>O:Ce showed an excitation peak at 326 nm and two excitation peaks, are each at 376 and 740 nm. The excitation peak at 326 nm and the emission peak at 370 nm are caused by the  $4f-5d$  transition. A second emission peak is observed at 700 nm; it also stems from the defect luminescence of SiMgAlLi<sub>2</sub>O:Ce [7].

### 3. Laser-induced emission spectrum at low temperature

A pulsed laser is used to measure the laser-induced emission spectrum of the SiMgAlLi<sub>2</sub>O:Ce and the SiMgAlLiF:Ce glasses at temperatures between 300 K and 10 K. Figure 6 shows that the light yields of both glass samples increase when the temperatures decreased.

These results are attributed to the thermal quenching effect [10]. Additional emission peaks are observed in the SiMgAlLi<sub>2</sub>O:Ce and SiMgAlLiF:Ce glasses. These peaks are produced by intrinsic defects in the glass samples [7]. Given the defect luminescence, the SiMgAlLi<sub>2</sub>O:Ce glass exhibits higher emission at a wavelength of 750 nm compared to the SiMgAlLiF glass. This indicates that the SiMgAlLi<sub>2</sub>O:Ce glass displays a higher quenching effect at room temperature than that of SiMgAlLiF:Ce glass does [11].

### 4. X-ray-induced emission spectrum

Figure 7 shows the X-ray-induced emission spectra of both glass samples. SiMgAlLiF:Ce exhibits an emission

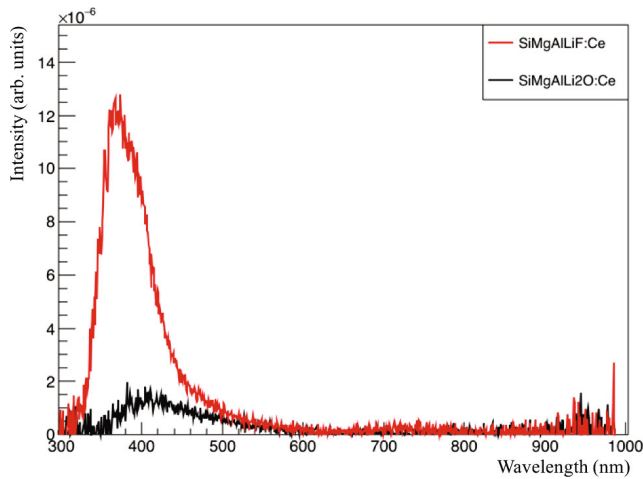


Fig. 7. (Color online) X-ray-induced emission spectra of SiMgAlLiF:Ce (red) and SiMgAlLi<sub>2</sub>O:Ce (black).

peak at 380 nm, and SiMgAlLi<sub>2</sub>O:Ce exhibits an emission peak at 410 nm. The intensity of the SiMgAlLiF:Ce emission exceeds that of SiMgAlLi<sub>2</sub>O:Ce. The emission peaks of both glass samples occur due to the  $4f$ - $5d$  transition of Ce<sup>3+</sup>. The intensity of the SiMgAlLiF:Ce emission exceeds that of the SiMgAlLi<sub>2</sub>O:Ce emission due to defect quenching [11].

### 5. Proton-induced emission spectrum

In order to measure the proton-induced emission spectrum of glass samples, we use the 100 MeV proton beam in KOMAC. Figure 8 shows the proton-induced emission spectrum of SiMgAlLiF:Ce. The proton-induced emission and X-ray-induced emission exhibit the same emission peaks. SiMgAlLi<sub>2</sub>O:Ce exhibits an extremely low light intensity; thus, an emission spectrum is absent.

## IV. CONCLUSIONS

SiMgAlLiF:Ce and SiMgAlLi<sub>2</sub>O:Ce glasses were fabricated using a melt-quenching technique. We measured laser-, photo-, X-ray-, and proton-induced luminescence spectra to examine the optical properties of the glass samples. Three decay times were found in both glass samples. The decay time of the major component was approximately 25 ns, but no significant differences in the decay times were observed with decreasing temperature. Both glass samples displayed an excitation peak and two emission peaks. The excitation and the emission peaks at 360 nm appeared to be due to the  $f$ - $d$  transition. The second emission peak at 700 nm in both glass samples and the peaks at 500 nm in the SiMgAlLi<sub>2</sub>O:Ce glass were caused by defect luminescence. The laser-induced

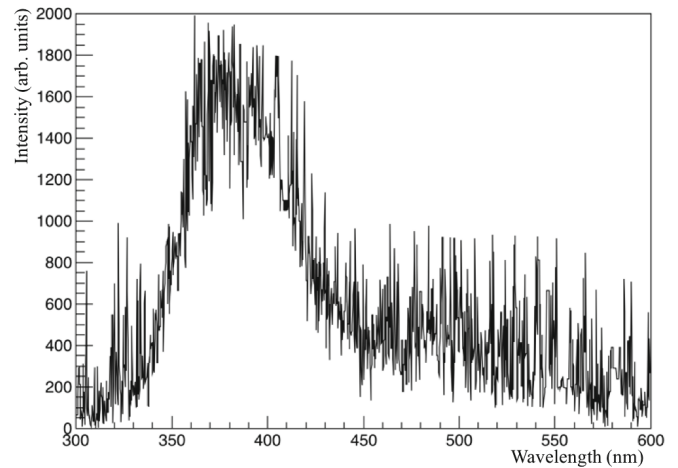


Fig. 8. Proton-induced emission spectrum of SiMgAlLiF:Ce glass.

emission spectra at low temperature were measured using the pulsed laser. The intensity of light increased with decreasing temperature in both glass samples. In the X-ray-induced emission spectra of glass samples, SiMgAlLiF:Ce exhibited an emission peak at 380 nm and SiMgAlLi<sub>2</sub>O:Ce exhibited an emission peak at 410 nm. The intensity of the SiMgAlLiF:Ce glass was approximately six times higher than that of the SiMgAlLi<sub>2</sub>O:Ce glass. The SiMgAlLi<sub>2</sub>O:Ce glass exhibited higher defect luminescence; thus, it displayed a higher defect quenching factor. Therefore, the SiMgAlLiF:Ce glass exhibited a light intensity higher than that of SiMgAlLi<sub>2</sub>O:Ce. We used a 100 MeV proton beam to study the proton-induced emission spectrum. A SiMgAlLiF:Ce displayed an emission peak at 380 nm. Proton-induced emission light of on SiMgAlLi<sub>2</sub>O:Ce was absent due to the extremely low light intensity. The glass scintillators can be easily fabricated in large sizes and various shapes. Both glass samples possessed fast decay times, emitted light in the visible region, and utilized Li for the host material. Therefore, both glass samples can be applied for high energy physics, nuclear physics, radiation monitoring, and neutron detection.

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