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# Luminescence decay evaluation of long-afterglow phosphors

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## ABSTRACT

In the current study, the phosphorescence decays of the long-afterglow materials of  $\text{SrAl}_2\text{O}_4:\text{Eu}$ ,  $\text{Dy}$ ,  $\text{Sr}_4\text{Al}_{14}\text{O}_{25}:\text{Eu}$ ,  $\text{Dy}$ ,  $\text{Sr}_2\text{MgSi}_2\text{O}_7:\text{Eu}$ ,  $\text{Dy}$ , and  $\text{ZnS}:\text{Cu}$ ,  $\text{Co}$  have been measured over long time (15 h to 16 h). The evaluation methods of luminescence decay are analyzed and summarized. The existing formulas are in accordance with the experimental decay data in the initial fast decay stage. However, the deviation is much larger and the accuracy is much lower in the slow decay stage. Therefore, the existing formulas cannot be used to accurately describe the luminescence decay of the samples with much longer afterglow time ( $> 80$  min). Moreover, certain formulas do not match the actual decay situation of long-afterglow luminescence. We suggest our solutions for this problem.

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## 1. Introduction

Long-afterglow refers to luminescence that can persist for hours after the excitation is stopped. Long-afterglow materials are widely used as night-vision materials in architectural decoration, transportation, military facilities, fire safety, and other daily necessities. To achieve practical levels of luminescence, there are two requirements for the materials: one is that the long-afterglow materials need to be easily excited by visible light; the other is that they must have sufficiently strong luminescence and long-afterglow time ( $> 10$  h) in order to absorb visible light during the daytime and luminescence during the entire night [1]. The afterglow luminescence decay time (LDT), as determined by the standard of safety signs and safe guidance systems (DIN 67510-1, ISO 16069), is the time that the afterglow material takes to decrease to the brightness of  $0.32 \text{ mcd/m}^2$  after the removal of excitation light. This is 100 times higher than the limit of light perception of the scotopic human eye ( $0.0032 \text{ mcd/m}^2$ ). Thus, the accurate measurement of LDT for long-afterglow materials is very important. However, some long-afterglow materials may maintain their luminescence for a few days without excitation light [2]. In such a case, it is very difficult to measure the full-life brightness and LDT of these materials.

In addition, a luminescence decay process is crucial to luminescence dynamics. However, little attention has been paid on the luminescence decay process since 1960s. According to the mechanism of long-afterglow luminescence, it is possible to obtain

the luminescence decay equation to extrapolate the LDT. However, the long lasting luminescence mechanism is not currently fully understood. A number of decay equations (Section 3) can be obtained according to different models. Some of the existing afterglow decay equations are listed in Table 1 which are very chaotic.

In our study of  $\text{Y}_2\text{O}_3:\text{Eu}$ ,  $\text{Mg}$ ,  $\text{Ti}$ , the afterglow decay cannot be described by Eqs. (8)–(10) shown in Section 3 [27]. Therefore, the decay formulas shown in Section 3 are used to study the afterglow decay of  $\text{SrAl}_2\text{O}_4:\text{Eu}$ ,  $\text{Dy}$ ,  $\text{Sr}_4\text{Al}_{14}\text{O}_{25}:\text{Eu}$ ,  $\text{Dy}$ ,  $\text{Sr}_2\text{MgSi}_2\text{O}_7:\text{Eu}$ ,  $\text{Dy}$  [28], and  $\text{ZnS}:\text{Cu}$ ,  $\text{Co}$  in this paper.

## 2. Experiment

Four types of commercial phosphors (PLO-8C, PLB-8C, SB-8C, and ZnS) that were obtained from Dalian Luming Light Company were analyzed by XRD and EDX. The phases were  $\text{SrAl}_2\text{O}_4:\text{Eu}$ ,  $\text{Dy}$ ,  $\text{Sr}_4\text{Al}_{14}\text{O}_{25}:\text{Eu}$ ,  $\text{Dy}$ ,  $\text{Sr}_2\text{MgSi}_2\text{O}_7:\text{Eu}$ ,  $\text{Dy}$ , and  $\text{ZnS}:\text{Cu}$ ,  $\text{Co}$ , respectively. The particle sizes were analyzed by using a Coulter LS100Q laser diffraction particle size analyzer. The average particle diameters were 26.12, 34.35, 37.65, and  $32.27 \mu\text{m}$ , with narrow particle size distributions. In the brightness characterization, fluctuations caused by scattering among particles were very slight ( $< 3\%$ ) because of the large particle size. Therefore, the brightness of the powder samples could be obtained directly. The photoluminescence spectra and afterglow intensity decay were investigated by using a Hitachi F-4500 fluorescence spectrophotometer equipped with a 150 W xenon lamp as the excitation source. The rapid decay data was recorded when the samples were first

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excited by sunlight for 5 min and then excited by the optimum excitation wavelength for 5 min (slit=10 nm) as shown in Fig. 2. Then, the samples were placed in a dark closet for 48 h before the brightness measurements. The afterglow brightness was measured by using a LMT photometer B-510 equipped with a D<sub>65</sub> standard light source of 1000 ± 10 lx in a dark room. The excitation time was 10 min based on DIN 67510 Part 1. The data recording interval was 1 min, and the entire measurement time was from 15 h to 16 h. The ambient temperature was from 22 °C to 24 °C. The afterglow luminance data of the samples were fitted by using the formulas shown in Section 3. The results are listed in Table 2, and the detailed fitting parameters are described in Tables s1–s15.

**Table 1**  
Existing afterglow decay equations.

Long afterglow luminescent materials	Afterglow decay equation in Section 3	Ref.
ZnS:Cu, SrAl <sub>2</sub> O <sub>4</sub> :Eu, and SrAl <sub>2</sub> O <sub>4</sub> :Eu, Dy	Eq. (17)	[3]
SrAl <sub>1.7</sub> B <sub>0.3</sub> O <sub>4</sub> :Eu, R (R=Nd, Dy)	Eq. (17)	[4]
SrAl <sub>2</sub> O <sub>4</sub> :Eu, Dy	Eq. (5)	[5]
SrAl <sub>2</sub> O <sub>4</sub> :Eu, Dy	Eq. (4)	[6]
BaAl <sub>2</sub> O <sub>4</sub> :Eu, Dy	Eq. (7)	[7]
Sr <sub>4</sub> Al <sub>14</sub> O <sub>25</sub> :Eu, Dy	Eq. (4)	[8]
Sr <sub>4</sub> Al <sub>14</sub> O <sub>25</sub> :Eu, Dy	Eq. (10)	[9]
SrAl <sub>4</sub> O <sub>7</sub> :Eu, Dy	Eq. (13)	[10]
Ca <sub>0.9</sub> Sr <sub>0.1</sub> S:Bi, Tm	Eq. (16)	[11]
CaAl <sub>2</sub> O <sub>4</sub> :Ce	Eq. (16)	[12]
M <sub>2</sub> MgSi <sub>2</sub> O <sub>7</sub> :Eu, Dy	Eq. (13), where I <sub>1</sub> =0	[13]
Sr <sub>2</sub> ZnSi <sub>2</sub> O <sub>7</sub> :Eu, Dy	Eq. (13), where I <sub>1</sub> =0	[14]
R <sub>3</sub> MgSi <sub>2</sub> O <sub>8</sub> :Eu, Dy (M: Ca, Sr, Ba)	Eq. (13), where I <sub>1</sub> =0	[15]
Ca <sub>8</sub> Mg(SiO <sub>4</sub> ) <sub>4</sub> Cl <sub>2</sub> :Eu	Eq. (9)	[16]
Ca <sub>8</sub> Zn(SiO <sub>4</sub> ) <sub>4</sub> Cl <sub>2</sub> :Eu	Eq. (4)	[17]
Y <sub>2</sub> O <sub>2</sub> S:Eu <sup>3+</sup> , Ti	Eq. (9)	[18]
Y <sub>2</sub> O <sub>2</sub> S:Eu, Mg, Ti	Eq. (10)	[19]
Y <sub>2</sub> O <sub>2</sub> S:Eu, Mg, Ti	Eq. (17)	[20]
Y <sub>2</sub> O <sub>2</sub> S:Sm	Eq. (4)	[21]
Ca <sub>2</sub> SnO <sub>4</sub> :Sm	Eq. (9)	[22]
CaGa <sub>2</sub> S <sub>4</sub> :Eu, Ho	Eq. (5)	[23]
Y <sub>3-x</sub> Mn <sub>x</sub> Al <sub>5-x</sub> Si <sub>x</sub> O <sub>12</sub>	Eq. (4)	[24]
CaWO <sub>4</sub> :Eu	Eq. (5)	[25]
Ca <sub>2</sub> SnO <sub>4</sub> :Pr	Eq. (9)	[26]

**Table 2**  
Fitting accuracy and LDT estimation of equations in Section 3.

Item	PLB-8C		PLO-8C		SB-8C		ZnS	
	R-square	LDT**	R-square	LDT	R-square	LDT	R-square	LDT
Eq. (3)	0.98097	40.4	0.97280	36.7	0.97280	34.5	0.95683	15.9
Eq. (4)	0.99870	133	0.99757	148	0.99757	135.5	0.99733	56
Eq. (5)	0.99984	330	0.99970	447	0.99970	380	0.99985	114
Eq. (6)	0.99998	730	0.99997	1070	0.99997	745	0.99997	145
Eq. (7)	1	1040	1	1490	1	935	1	155
Eq. (8)	0.98224	∞	0.97533	∞	0.97533	∞	0.96540	∞
Eq. (9)	0.99896	∞	0.99820	∞	0.99820	∞	0.99837	∞
Eq. (10)	0.99990	∞	0.99983	∞	0.99983	∞	0.99993	∞
Eq. (11)	0.98169	∞	0.97318	∞	0.97533	∞	0.96518	∞
Eq. (12)	0.99890	∞	0.99830	∞	0.99820	∞	0.99836	∞
Eq. (13)	0.99990	∞	0.99986	∞	0.99983	∞	0.99993	∞
Eq. (15)	0.99571	540	0.99108	520	0.99108	262	0.99338	94.5
Eq. (16)	0.99977	6000	0.99965	17,500	0.99965	4000	0.99984	300
Eq. (17)	0.98809	155,000	0.99257	116,000	0.99257	27,000	0.99786	520
Eq. (18)	0.99991	1780	1	3800	1	1200	0.99783	169
ALDT*	-	920(1.00)	-	910(2.76)	-	970(0.41)	-	157(0.32)

\* ALDT: Actual measured decay time (min) the sample takes to decrease to the brightness value (mcd/m<sup>2</sup>) in the parentheses.

\*\* LDT: The afterglow luminescence decay time (min) the sample takes to decrease to a brightness of 0.32 mcd/m<sup>2</sup> by extrapolation of the decay equation.

### 3. Long-afterglow luminescence decay mechanism

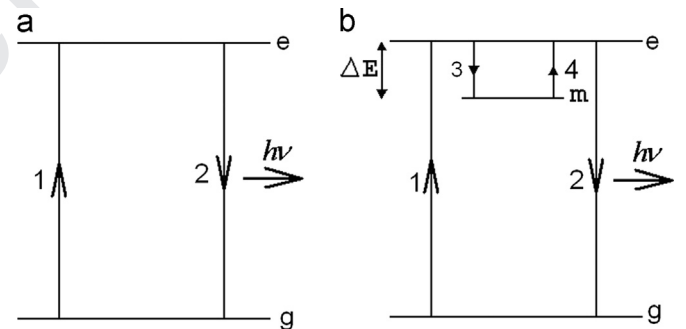
Fig. 1 shows a schematic of the fluorescence and long-afterglow mechanism model [1]. An electron is promoted from the ground state (g) to an excited state (e) when the material is excited by external radiation (process 1). Then, the transition from e to g state generates fluorescence (process 2). The interval time is very short between the processes 1 and 2 (approximately 10<sup>-8</sup> s). The entire process is independent of temperature (Fig. 1a). However, if metastable state trap levels exist (m), the electrons at the e state will be captured and stored by the m state (process 3). Then, the bound electrons gradually leap out of the m state due to the thermal disturbances (process 4), yielding afterglow luminescence (process 2). Therefore, the interval time will be extended between the processes 1 and 2, and the delay time is determined by the time taken by electrons to stay in the m state. The delay time can be as short as a few seconds or it can be up to tens of hours (Fig. 1b).

According to thermodynamic theory, the average capture time  $\tau$  depends on the trap depth  $\Delta E$  and temperature  $T$  [29,30]:

$$\tau = s^{-1} e^{\Delta E/kT}, \quad (1)$$

where  $s$  is the frequency factor for electron detrapping and  $k$  is the Boltzmann constant.

If the recaptured probability of the electrons released from the m state is negligible, the emission intensity  $I(t)$  is proportional to



**Fig. 1.** Schematic of fluorescence (a) and long-afterglow mechanism model (b).

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