

## 초청총설논문

### 플라즈마 표시판용 형광체

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## Phosphors for Plasma Display Panels

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**요약.** 플라즈마 표시판용으로 사용되거나 제안 또는 알려진 형광체들의 들뜸 및 발광 특성을 살펴 보았다. 각 형광체의 발광 효율이 임자 결정과 활성체의 종류에 따라 어떻게 달라지는지 알아 보았고 실용성 있는 형광체의 개발 방향을 생각하였다.

**ABSTRACT.** Excitation and emission characteristics were reviewed for phosphors which were reported, applied, or suggested for the plasma display panel (PDP). Correlation of luminescence characteristics to the host crystal structure and the activator of the phosphor was explained. Improvements of the PDP phosphor for the practicality were considered.

### INTRODUCTION<sup>1-4</sup>

High definition television (HDTV) sets of large screen cathode ray tubes (CRT) are too bulky and take up too much space for the wide spread acceptance. Thus flat panel displays (FPD) such as the liquid crystal display (LCD), the electroluminescence display (ELD) and the plasma display panel (PDP), have a competitive edge in the large screen display market.

The LCD is one of the most advanced technologies of FPDs. Rather restricted viewing angle of the LCD, which was one of the weaknesses of the LCD, has been relieved by employing the thin film transistor (TFT) active matrix system. However, drawbacks such as the assembly difficulty of the larger screen and the slow response speed are main obstacles for the commercialization of LCDs with screens larger than 30 inches. The ELD also is fairly advanced but full color ELDs are not available commercially. Only monochrome and mul-

ticolor ELDs are available for portable computers and medical instruments. The PDP would be the most promising FPD for the wall-hanging HDTV because the larger sized PDP could be made relatively easily. And it offers a fast response, a wide viewing angle, a low energy consumption and other advantages.

In PDPs, three primary color phosphors are excited by the vacuum ultraviolet (vuv) radiation from an inert gas plasma, which imposes a unique requirements to the phosphors. In this paper we will discuss characteristics of phosphors and some guidelines on designing new materials for PDP.

### PHOSPHORS FOR PDPs

The gas discharge FPD, *i.e.* PDP, was first suggested in 1964 by D. L. Bitzer at University of Illinois.<sup>5</sup> The general structure of a PDP cell is shown in Fig. 1.<sup>6</sup> Phosphors are excited by more energetic vuv radiation of wavelengths of 130 nm.

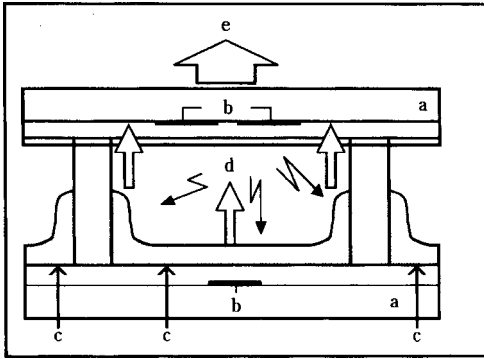


Fig. 1. A typical cell structure of a PDP. a: glass substrate, b: electrodes, c: R, G, or B phosphor, d: plasma discharge, e: visible light output.

147 nm and 172 nm from Xe and Xe-He discharges. The vuv radiation output of the plasma source depends on the gas constituent, composition and the pressure as shown in Fig. 2.<sup>7</sup> The energy conversion efficiency of the vuv in the range of 7 to 12 eV into the visible is generally higher at lower energies. Thus the xenon is the preferred constituent for the PDP.

PDP phosphors are strongly resemble fluorescent lamp phosphors. Some of the phosphors are listed in Table 1 along with standard color coordinates of the NTSC red, green, and blue phosphors.<sup>8-17</sup> From the Table 1, fairly low (near 1%) radiant efficiencies for these phosphors are no-

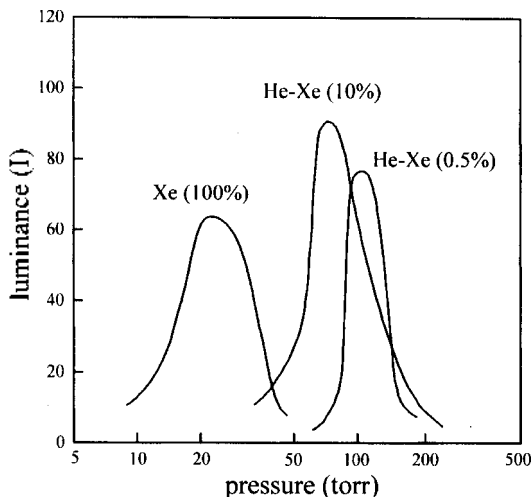


Fig. 2. Pressure dependence of the plasma vuv radiation.

Table 1. Characteristics of some phosphors

Phosphor	CIE coordinate		Relative radiant efficiency	$\tau_{1/2}$ (ms)	Brightness (cd/m <sup>2</sup> )
	x	y			
NTSC Red	0.67	0.33			
Y <sub>2</sub> O <sub>3</sub> :Eu	0.648	0.347	0.67	1.3	62
(Y,Gd)BO <sub>3</sub> :Eu	0.641	0.356	1.2	4.3	
YBO <sub>3</sub> :Eu		0.35	1.0		
GdBO <sub>3</sub> :Eu	0.65	0.36	0.94		
LuBO <sub>3</sub> :Eu	0.64	0.37	0.74		
ScBO <sub>3</sub> :Eu	0.63	0.39	0.94		
Y <sub>2</sub> SiO <sub>5</sub> :Eu	0.61	0.34	0.67		
Y <sub>3</sub> Al <sub>5</sub> O <sub>12</sub> :Eu	0.66	0.37	0.47		
Zn <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub> :Mn	0.63	0.33	0.34	15.1	
NTSC Green	0.21	0.71			
Zn <sub>2</sub> SiO <sub>4</sub> :Mn	0.242	0.708	1.0	11.9	365
BaAl <sub>12</sub> O <sub>19</sub> :Mn	0.182	0.732	1.1	7.1	
SrAl <sub>12</sub> O <sub>19</sub> :Mn	0.16	0.75	0.62		
CaAl <sub>12</sub> O <sub>19</sub> :Mn	0.15	0.75	0.34		
ZnAl <sub>12</sub> O <sub>19</sub> :Mn	0.17	0.74	0.54		
BaMgAl <sub>14</sub> O <sub>23</sub> :Mn	0.15	0.73	0.92		
YBO <sub>3</sub> :Tb	0.33	0.61	1.1		
LuBO <sub>3</sub> :Tb	0.33	0.61	1.1		
GdBO <sub>3</sub> :Tb	0.33	0.61	0.53		
ScBO <sub>3</sub> :Tb	0.35	0.60	0.36		
Sr <sub>2</sub> Si <sub>9</sub> O <sub>6</sub> Cl <sub>4</sub> :Eu	0.14	0.33	1.3		
NTSC Blue	0.14	0.08			
BaMgAl <sub>10</sub> O <sub>17</sub> :Eu	0.147	0.067			
BaMgAl <sub>14</sub> O <sub>23</sub> :Eu	0.142	0.087	1.6	< 1	
Y <sub>2</sub> SiO <sub>5</sub> :Ce	0.16	0.09	1.1	< 1	
CaWO <sub>4</sub> :Pb	0.17	0.17	0.74		51

ticeable. Various hosts and activators have been studied but phosphors with better performances should be found for a competitive PDP realization. Until recently, the main PDP phosphors are Y<sub>2</sub>O<sub>3</sub>:Eu and (Y,Gd)BO<sub>3</sub>:Eu for red, Zn<sub>2</sub>SiO<sub>4</sub>:Mn and BaAl<sub>12</sub>O<sub>19</sub>:Mn for green, and BaMgAl<sub>14</sub>O<sub>23</sub>:Eu and BaMgAl<sub>10</sub>O<sub>17</sub>:Eu for blue. Excitation and emission spectra of these tricolor phosphors are shown in Fig. 3a and b.

As listed in Table 1, decay times of (Y,Gd)BO<sub>3</sub>:Eu<sup>3+</sup> and BaAl<sub>12</sub>O<sub>19</sub>:Mn are somewhat longer, but those of Zn<sub>3</sub>(PO<sub>4</sub>)<sub>2</sub>:Mn and Zn<sub>2</sub>SiO<sub>4</sub>:Mn are too long for the practical application. Since decay times of red and green phosphors are longer to exploit the fast response of the PDP, they should be improved. The blue phosphor BaMgAl<sub>14</sub>O<sub>23</sub>:Eu<sup>2+</sup>

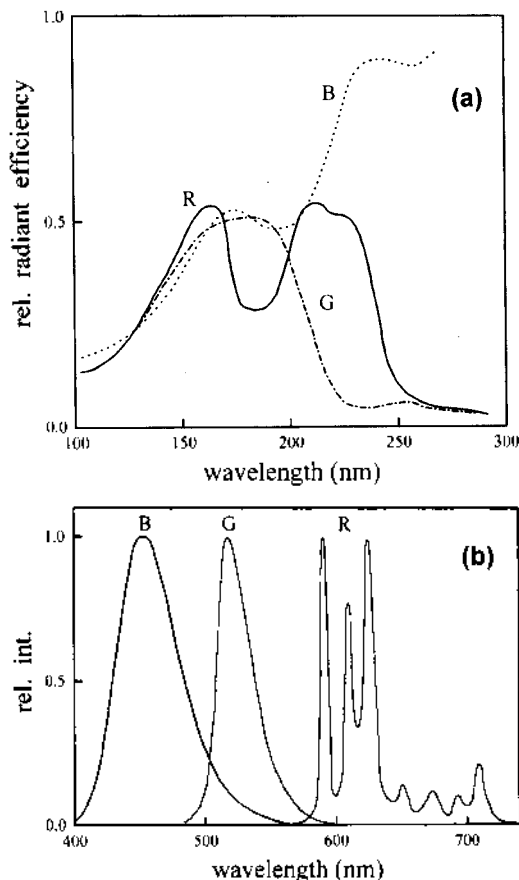


Fig. 3. (a) Excitation spectra of R, G, and B phosphors. (b) Emission spectra of R, G, and B phosphors. R:  $(Y,Gd)BO_3:Eu^{3+}$ , G:  $BaAl_{12}O_{19}:Mn$ , B:  $BaMgAl_{14}O_{23}:Eu^{2+}$ .

has a fast decay time which is obscured by the plasma discharge decay, thus accurate measurement is not easy.

### HOST SENSITIZATION

When phosphors are excited by vuv, the influence of the host lattice is very important. For example, the emission spectrum of  $(Y,Gd)BO_3:Eu^{3+}$  peaks at about 595 nm which is shorter than the peak emission wavelength of  $Y_2O_3:Eu$ . However, its performance on excitation in the vuv region is better than that of  $Y_2O_3:Eu$  as shown in Fig. 4. The host excitation band of  $(Y,Gd)BO_3:Eu^{3+}$  appears at near 160 nm.

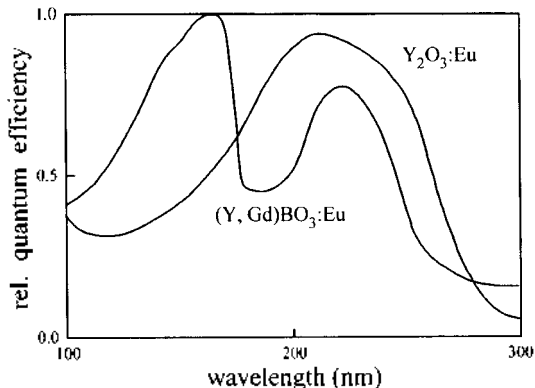


Fig. 4. Excitation spectra of  $Y_2O_3:Eu$  and  $(Y,Gd)BO_3:Eu$ .

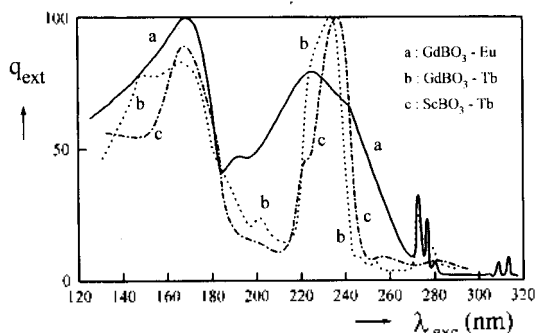


Fig. 5. Excitation spectra of  $GdBO_3:Eu$ (a),  $GdBO_3:Tb$ (b) and  $ScBO_3:Tb$ (c).

Comparing the  $Eu^{3+}$ -activated and  $Tb^{3+}$ -activated Y borate with Gd borate and Sc borate,<sup>18</sup> we could note that they all show a peak in the excitation spectrum near 160 nm (see Fig. 5 and Fig. 6) with slight differences in the peak intensities. The  $Eu^{3+}$ -activated borates, in particular, show high quantum efficiencies (see Table 1).

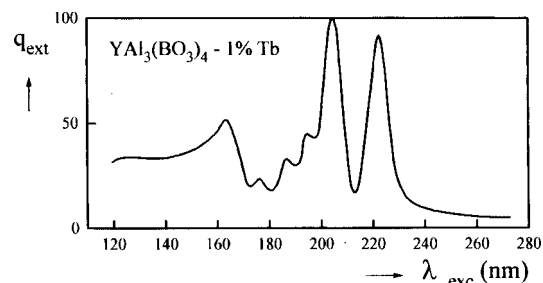


Fig. 6. Excitation spectrum of  $YAl_3(BO_4)_4:Tb$ .

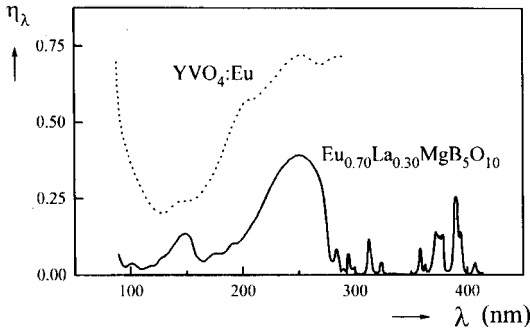


Fig. 7. Excitation spectrum of  $\text{Eu}_{0.70}\text{La}_{0.30}\text{MgB}_5\text{O}_{10}$  (—) and  $\text{YVO}_4:\text{Eu}$  (---).

Because peaks near 160 nm are present both in  $\text{Eu}^{3+}$ - and in  $\text{Tb}^{3+}$ -activated borates, the absorption in this region is most probably due to the host lattice, i.e., in the  $\text{BO}_3$  groups. The excitation spectrum  $\text{Eu}_{0.70}\text{La}_{0.30}\text{MgB}_5\text{O}_{14}$  shows the host sensitization band of  $\text{BO}_3$  groups at around 150 nm as in Fig. 7.<sup>19</sup>

The excitation spectrum of  $\text{Tb}_x\text{Y}_{1-x}\text{PO}_4$  is shown in Fig. 8. Hoshina<sup>20</sup> reported that peak A is a spin-forbidden transition and B is a spin-allowed 4f-5d transition. It is found that in  $\text{Tb}_x\text{Y}_{1-x}\text{PO}_4$  peaks C and A have one type of concentration dependence, while peaks B and D exhibit another type of concentration dependence. The peak C can be as-

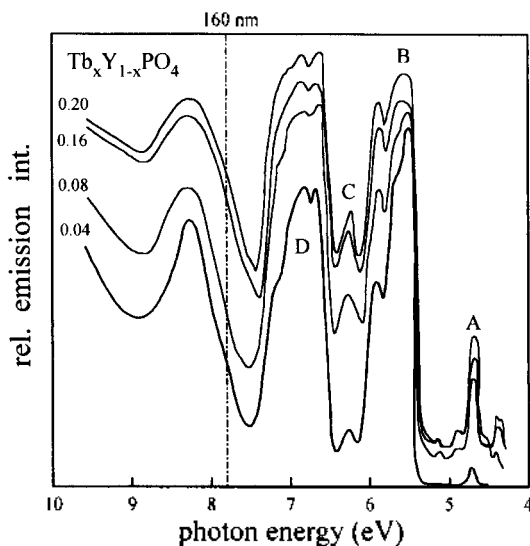


Fig. 8. Excitation spectra of  $\text{Tb}_x\text{Y}_{1-x}\text{PO}_4$ .

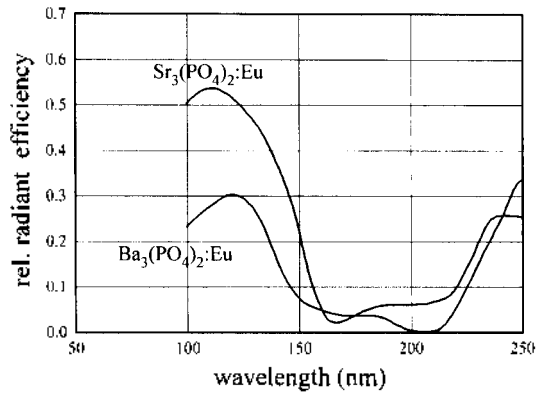


Fig. 9. Excitation spectra of  $\text{Sr}_3(\text{PO}_4)_2:\text{Eu}$  and  $\text{Ba}_3(\text{PO}_4)_2:\text{Eu}$ .

cribed to a spin-forbidden transition as A, and the peak D to a spin-allowed transition as B. The band with a peak at 150~160 nm is assumed to be a host sensitization band of the  $\text{PO}_4^{3-}$  group. The fact that this excitation band is found for  $\text{YPO}_4$  host can be interpreted as a result of an efficient energy transfer from  $\text{PO}_4^{3-}$  group to the activator.

Excitation spectra of  $\text{Sr}_3(\text{PO}_4)_2:\text{Eu}$  and  $\text{Ba}_3(\text{PO}_4)_2:\text{Eu}$  are shown in Fig. 9.<sup>21</sup> Excitation band with a peak around 125 nm is observed in the excitation spectrum of the  $(\text{PO}_4)_2^{6-}$ . While 4f-5d transitions of  $\text{Tb}^{3+}$  in  $\text{TbP}_5\text{O}_{14}$  are nearly the same as in other type of phosphates as shown in Fig. 10, but the

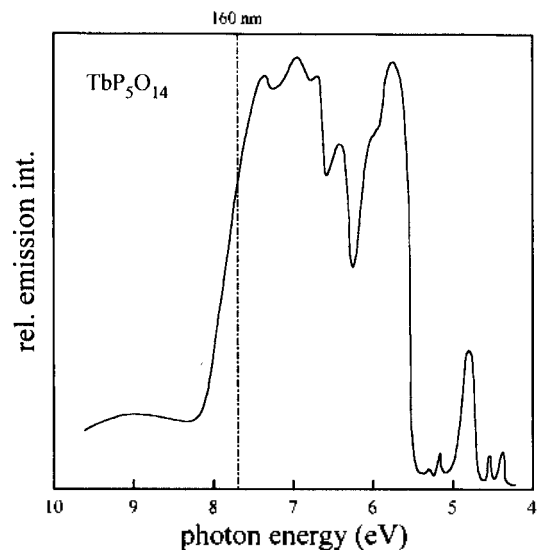


Fig. 10. Excitation spectrum of  $\text{TbP}_5\text{O}_{14}$ .

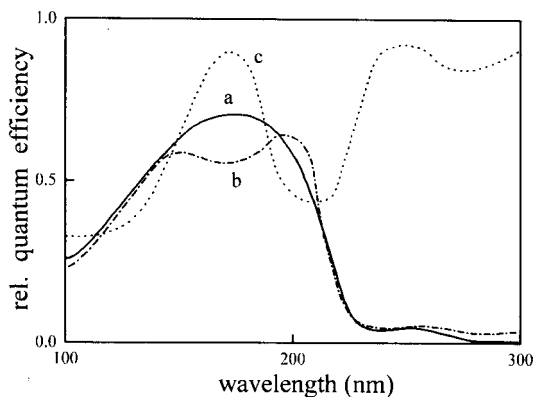


Fig. 11. Excitation spectra of phosphors. (a)  $\text{BaAl}_{12}\text{O}_{19}:\text{Mn}$ , (b)  $\text{BaMgAl}_{14}\text{O}_{23}:\text{Mn}$ , (c)  $\text{BaMgAl}_{14}\text{O}_{23}:\text{Eu}$ .

host sensitization band of  $\text{P}_5\text{O}_{14}^{3-}$  groups shifts to longer wavelength of near 135 nm compared to di-phosphates.

All the excitation spectra of  $\text{BaAl}_{12}\text{O}_{19}:\text{Mn}$  with  $\text{BaMgAl}_{14}\text{O}_{23}:\text{Mn}$  and  $\text{BaMgAl}_{14}\text{O}_{23}:\text{Eu}$  give a weak band near 175 nm as shown in Fig. 11. This can be attributed to the host sensitization band of aluminates at this range.

As shown in Fig. 12,  $\text{CaO}:\text{Eu}^{3+}$  gives an absorption band at 205 nm which is presumed<sup>22</sup> to be a host excitation band due to a transition from the valence band to the conduction band *i.e.*, the excitation energy is absorbed by the host and transferred to the activator in the relaxation process. The 254-nm band is the well known charge transfer band in  $\text{Eu-O}$  *i.e.*, the electron transfer from oxygen to  $\text{Eu}^{3+}$  ion.

In the oxyfluoride system  $(\text{CaF}_2, \text{CaO}):\text{Eu}^{3+}$  a broad band merged with additional features from  $\text{CaF}_2:\text{Eu}^{3+}$  in the higher energy region appears with increased vuv absorption as shown in Fig. 13. By comparing the excitation spectrum of  $\text{CaO}:\text{Eu}^{3+}$  with that of  $(\text{CaF}_2, \text{CaO}):\text{Eu}^{3+}$  it is quite easy to notice that the introduction of F ions in the host lattice shifts all the features to a higher energy region. This is in full agreement with the fact that the charge transfer occurs at higher energy in  $\text{Eu-F}$  system than in the oxygen dominating lattice, according to the respective electronegativity of the ligand.

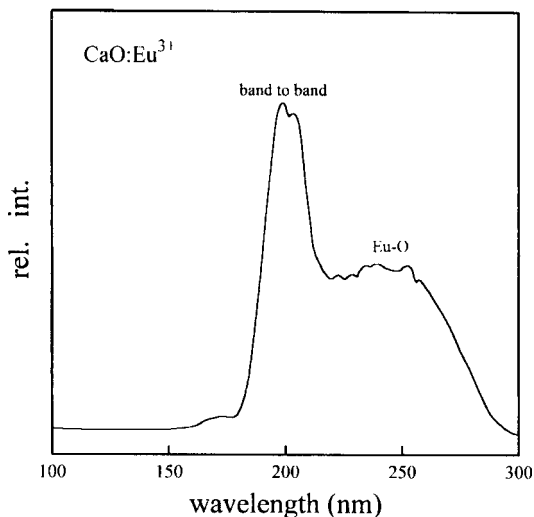


Fig. 12. Excitation spectrum of  $\text{CaO}:\text{Eu}^{3+}$ .

In order to check the fluoride behavior, the  $\text{LaF}_3:\text{Eu}^{3+}$  phosphor was investigated as shown in Fig. 14. The features on the excitation spectrum are clearly identifiable: the charge transfer (CT) band occurs at 170 nm and the band to band transition at 125 nm. The host sensitization bands in rare earth ion activated  $\text{LiYF}_4$ ,  $\text{LaF}_3$  and  $\text{YF}_3$  are reported to be at near 120 nm.<sup>23-28</sup>

As explained above the host sensitization band in different hosts have different position, and the host sensitization band positions are: oxide ( $\text{CaO}:\text{Eu}$ , ~

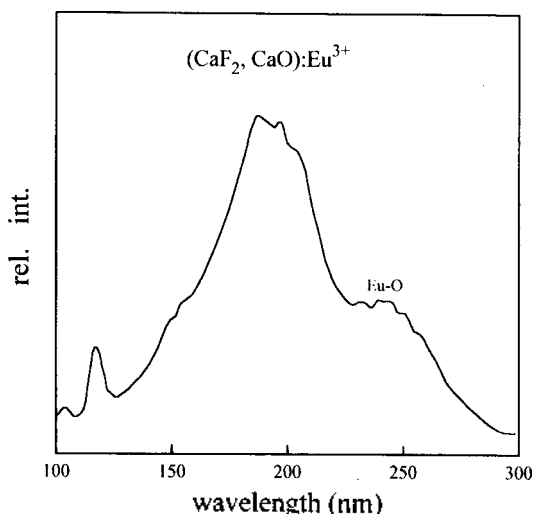


Fig. 13. Excitation spectrum of  $(\text{CaF}_2, \text{CaO}):\text{Eu}^{3+}$ .

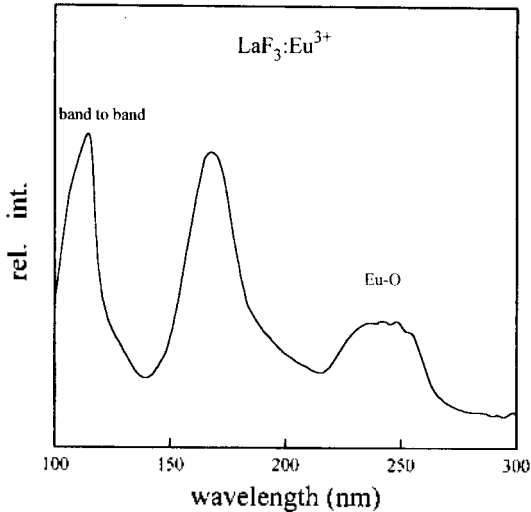


Fig. 14. Excitation spectrum of  $\text{LaF}_3:\text{Eu}^{3+}$ .

200 nm) > multialuminate ( $\text{BaMgAl}_{14}\text{O}_{23}:\text{Eu}$ , ~175 nm) > borate ( $\text{YBO}_3:\text{Eu}$ , 150~170 nm) > vanadate ( $\text{YVO}_4:\text{Eu}$ , ~150 nm) = phosphate ( $\text{YPO}_4:\text{Eu}$ , ~150 nm) > pentaphosphate ( $\text{TbP}_5\text{O}_{14}$ , ~135 nm) > diphosphate ( $\text{Ba}_3(\text{PO}_4)_2:\text{Eu}$ , ~125 nm) > fluoride ( $\text{LaF}_3:\text{Eu}$  or  $\text{LiYF}_4:\text{Eu}$ , ~120 nm). These results would be the basis of designing new materials for PDP phosphors.

### THE EFFECT OF CATION (rare-earth ion)

The energy of the first f-d transition in trivalent lanthanide ion and band gap energy of the three different fluorides<sup>29</sup> are summarized in Table 2. Table 2 shows that the energy level of rare earth trivalent ion are spreaded over the range of vuv and the phosphors with these hosts are excited by the vuv.

When  $\text{Sm}^{3+}$ ,  $\text{Eu}^{3+}$  or  $\text{Gd}^{3+}$  are doped in  $\text{YPO}_4$ , the host sensitization band of  $\text{YPO}_4$  stays unchanged in the excitation spectra at around 152 nm as shown in Fig. 15.<sup>30</sup>

Excitation spectra of  $\text{LaPO}_4$ ,  $\text{GdPO}_4$ ,  $\text{YPO}_4$  and  $\text{LuPO}_4$  activated with 0.5~2 mol% of  $\text{Eu}^{3+}$  are shown in Fig. 16. It is noted that  $\text{YPO}_4$  and  $\text{LuPO}_4$  show the similar spectra and so do  $\text{LaPO}_4$  and  $\text{GdPO}_4$ . Peak positions of the host sensitization band of  $\text{LaPO}_4$  (159 nm) and  $\text{GdPO}_4$  (160 nm) are

Table 2. The first f-d transition energy of lanthanides in fluoride hosts

Element	No. of f e	$E_{f-d}^a$ of tri-valent RE ions in					
		$\text{LiYF}_4$		$\text{YF}_3$		$\text{LaF}_3$	
		(eV)	(nm)	(eV)	(nm)	(eV)	(nm)
Ce	1	4.19	296	4.9	253	4.96	250
Pr	2	5.85	212	6.59	188	6.59	188
Nd	3	7.1	175	7.33	169	7.65	162
Pm	4	-	-	-	-	-	-
Sm	5	-	-	-	-	-	-
Eu	6	8.67	143	9.31	133	9.5	130.5
Gd	7	-	255	-	-	-	-
Tb	8	4.86	191	5.82	213	6.13	202
Dy	9	6.48	154	8	155	8	155
Ho	10	8.05	154	8.85	140	9.2	135
Er	11	8.05	154	8.38	148	8.65	143
Tm	12	7.95	156	8.33	149	8.65	143
Yb	13	-	-	-	-	-	-
$E_{\text{band}}^b$		10.55	117.5	10.53	118	10.51	118
$E_{\text{charge}}^c$		7.9	157	7.88	157	7	177

<sup>a</sup> $E_{f-d}$ : the first f-d transition energy. <sup>b</sup> $E_{\text{band}}$ : band to band transition energy of the host. <sup>c</sup> $E_{\text{charge}}$ : charge transfer energy of the host.

very close to each other and are shifted to longer wavelengths than that of  $\text{YPO}_4$  (152 nm) and  $\text{LuPO}_4$  (145 nm, 151 nm). Bandwidths of the host sensitization of the former two ions are larger than those of the latter two due to tailings in the short wavelength side.  $\text{YPO}_4$  and  $\text{LuPO}_4$  have the tetrag-

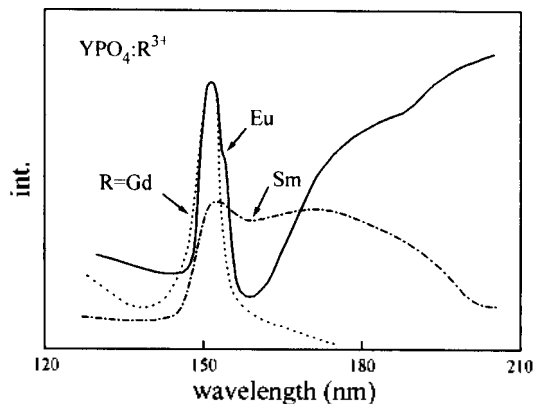


Fig. 15. Excitation spectra of  $\text{YPO}_4:\text{R}^{3+}$  (0.5~2 mol%). R=Gd (310 nm emission), Sm (610 nm emission), and Eu (620 nm emission).

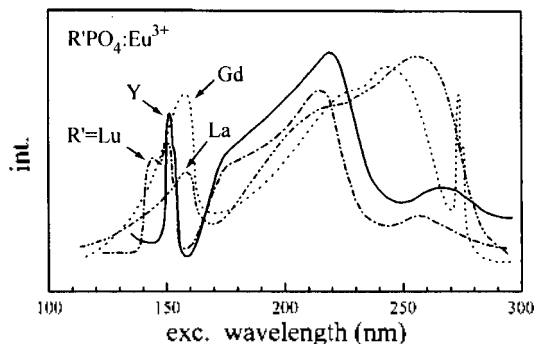


Fig. 16. Excitation spectra of  $R'PO_4:Eu^{3+}$  (0.5~2 mol%), where  $R=La, Gd, Lu$  and  $Y$ .

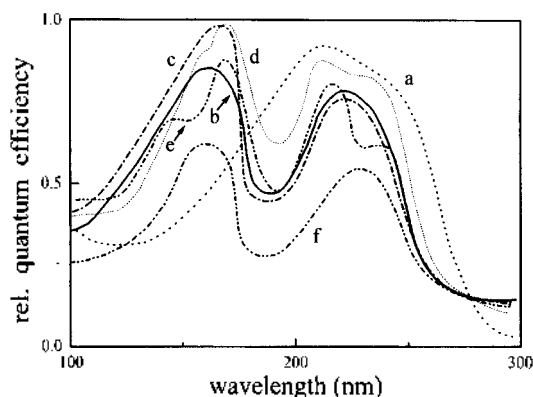


Fig. 17. Excitation spectra of red emitting phosphors. (a)  $Y_2O_3:Eu$ , (b)  $YBO_3:Eu$ , (c)  $(Y,Gd)BO_3:Eu$ , (d)  $GdBO_3:Eu$ , (e)  $ScBO_3:Eu$ , (f)  $LuBO_3:Eu$ .

onal zircon structure ( $D_{4h}$ ),<sup>19</sup> while  $LaPO_4$  and  $GdPO_4$  have the monoclinic monazite structure ( $C_{2h}$ ).<sup>5</sup> Thus the shift of the host sensitization bands as well as the CT bands could be considered to reflect the difference in the crystal structure of the hosts.

Excitation spectra of red-emitting phosphors for different rare earth ions in  $LnBO_3:Eu^{3+}$  are shown in Fig. 17. As the radius  $Ln^{3+}$  increases, the host sensitization band moves to longer wavelengths. The same trend could be observed as shown in Fig. 18 in the system of  $LnBO_3:Tb^{3+}$  ( $Ln=Gd, Y$ , or  $Lu$ ) *i.e.*, the order of the host sensitization bands are:  $\lambda_{exc}(GdBO_3:Tb) > \lambda_{exc}(YBO_3:Tb) > \lambda_{exc}(LuBO_3:Tb)$ .

## PHOSPHORS STABILITY

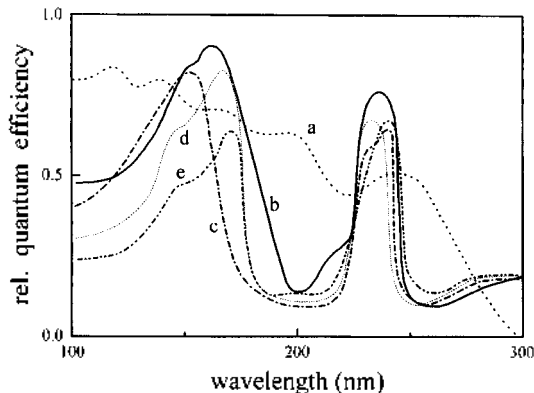


Fig. 18. Excitation spectra of green emitting phosphors. (a)  $Zn_2SiO_4:Mn$ , (b)  $YBO_3:Tb$ , (c)  $LuBO_3:Tb$ , (d)  $GdBO_3:Tb$ , and (e)  $ScBO_3:Tb$ .

The deterioration of phosphors is caused by the heat treatment during the panel fabrication process, *i.e.*, burning off the binder from the phosphor screen, sealing the glass frit, and exhausting the panel. As shown in Table 3,  $BaAl_{12}O_{19}:Mn$  exhibits almost no loss in efficiency after baking in air (460 °C, 20 min.), while  $(Y,Gd)BO_3:Eu$  and  $BaMgAl_{14}O_{23}:Eu$  display a decrease of 10 % in efficiency.

The temperature dependence of relative luminance of some phosphors are shown in Fig. 19. At higher temperatures,  $(Y,Gd)BO_3:Eu$  and  $BaAl_{12}O_{19}:Mn$  give somewhat higher efficiencies and  $BaMgAl_{14}O_{23}:Eu$  exceeds  $CaWO_4$  and  $Zn_2SiO_4:Mn$ .

PDP phosphors are exposed to the vuv of high energy and high intensity. Thus the radiation stability of phosphors should be considered seriously. The color center and surface defects of the phosphor caused by the radiation will decrease luminance efficiency. Practical results indicate that

Table 3. Phosphor deterioration upon baking

Phosphor	Brightness retention*
$(Y,Gd)BO_3:Eu$	0.91
$BaAl_{12}O_{19}:Mn$	0.99
$Zn_2SiO_4:Mn$	0.96
$BaMgAl_{14}O_{23}:Eu$	0.88

\*normalized to the value before baking.

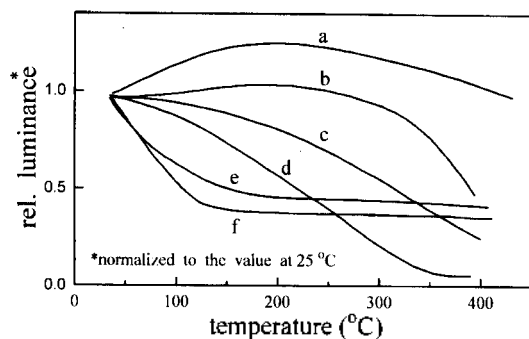


Fig. 19. Temperature dependence of several phosphors. (a)  $(Y,Gd)BO_3:Eu^{2+}$ , (b)  $BaAl_{12}O_{19}:Mn$ , (c)  $BaMgAl_{14}O_{23}:Eu^{2+}$ , (d)  $Zn_2SiO_4:Mn$ , (e)  $CaWO_4$ , (f)  $ZnS:Ag$ .

the  $Eu^{2+}$  in phosphors is unstable, because it could be oxidized to  $Eu^{3+}$  under the vuv radiation.

The performance of the state-of-the-art commercial PDP phosphors are still not good enough for the practical application. For the wide spread acceptance of the PDP new phosphors should be developed in the near future.

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