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Investigation on the conductivity-dependent performance in low voltage cathodoluminescence

Chunyu Shang,* Jinxian Zhao, Xiuqin Wang, Hongyang Xia and Hui Kang

With the increasing of applied current density in the low voltage cathodoluminescence, the exciting power tends to saturate, causing the saturation behavior of electron-hole generation rate in the phosphor layer. Meanwhile, ground-state depletion would emerge for the activators owing to the increased exciting power and decreased average penetration depth of incident electrons in the phosphor layer, causing the decrease of energy transfer probability of e-h pairs exciting ground-state activators. In addition, the radiative transition probability of excited activators would be decreased due to the increasing of temperature. In view of these key factors, the efficiency decrease in cathodoluminescence is the inevitable result. To restrain the efficiency decrease so as to improve the performance in low voltage cathodoluminescence, the conductivity of the phosphor material were to be improved. By introducing conductive component, improving the conductivity of the phosphor material, the performance in low voltage cathodoluminescence was effectively improved.

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1. Introduction

Nowadays, several technologies have been developed for the flat panel displays (FPDs), and FPDs with improved display quality and enhanced energy efficiency are strongly required. Of these technologies, field emission displays (FEDs) have been developed as one of the most promising next generation flat panel displays due to their potential advantages. The basis of an FED device is an array of field emitting cathodes aligned opposite to an array of phosphor anodes. The operation principle of FEDs is similar to that of conventional cathode ray tubes (CRTs), in which the fundamental mechanism is the cathodoluminescence.[1–6] Specially, FED should operate at a lower voltage (< 4kV), hence a larger current density (10-100 μA/cm^2) is required to guarantee a high output power.[7] However, operated at a low voltage, the efficiency decrease in cathodoluminescence would be typically exhibit with the increasing of current density, degrading the performance in FED. This is one of the most serious impediments hindering the development in FED. Consequently, it is essential to investigate the mechanisms in the efficiency decrease with the increasing of current density in low voltage cathodoluminescence. In our research work, based on comprehensive investigations, we have revealed in several aspects that the conductivity of the phosphor material has profound influences on the performance in low voltage cathodoluminescence, and a new level of understanding has been achieved. As a result, the ultimate method improving the performance in low voltage cathodoluminescence is presented. Clearly, as the prerequisite for further experimental investigations, our present research work in the low voltage cathodoluminescence is of significant importance.

2. Experimental

Y_2O_3:Eu^{3+} phosphor was prepared in glycine-nitrate solution combustion process.[8] Lanthanide nitrate ($Y(NO_3)_3 \cdot 6H_2O$, $Eu(NO_3)_3 \cdot 6H_2O$) and glycine ($C_2H_5NO_2$) were dissolved in distilled water (glycine-to-nitrate ratio was set to be 1.7) to form the precursor solution. The precursor solution was concentrated by heating at 260ºC until spontaneous combustion occurred. After being heat-treated at 700ºC for an hour, Y_2O_3:Eu^{3+} phosphor was formed.

Y_2O_3:Eu^{3+} phosphor was attached with In_2O_3 by hydrolysis of indium isopropoxide ($In[OCH(CH_3)_2]_3$) in the sol-gel process. Y_2O_3:Eu^{3+} powder phosphor, distilled water and isopropyl alcohol were added into the alkoxide solution of In. During churning of these reactants at 80ºC, hydrolysis and condensation occurred. After drying the solution and heated at 300ºC for 3 hours, In_2O_3-introduced Y_2O_3:Eu^{3+} phosphor was formed.

In the resistivity measurement, each of the phosphor samples was made into a cylindrical with diameter $2r$ = 13mm and thickness d = 2mm under a pressure of 1.5×10^2 Mpa. Electrodes were made by Al depositions on both sides of each sample. The resistance

R was measured with a 769YP-24B megohmmeter and the conductivity was calculated by the formula, $\sigma = d/\pi \cdot r^2 \cdot R$.

The cathodoluminescence measurements were performed in an ultra-high-vacuum chamber (<10^{-8} Torr), where the samples were excited by an electron beam at different voltages and different current densities, and the emission spectra were recorded by using an F-7000 spectrophotometer.

3. Results and discussion

3.1 Related mechanisms in the phosphor layer in low voltage cathodoluminescence

In the low voltage cathodoluminescence, when the anode-cathode current, which is controlled by the gate voltage, is applied, the electrons would be emitted from the cathode and accelerated (by the anode-cathode voltage) flying to the anode, exciting the phosphor layer on the anode surface. An incident electron would then be scattered constantly in the phosphor layer until its kinetic energy is exhausted. Initially, the current in the phosphor layer to the anode should be lower than that emitted from the cathode, causing the accumulations of negative charges in the phosphor layer. According to the related physical principles, with the negative charges accumulating in phosphor layer, positive charges, which are the corresponding induced charges of the same quantity, would be accumulated simultaneously on the anode surface. With the charge accumulations, the electric field in the phosphor layer is set up, promoting an intrinsic current in the phosphor layer. This process would continue until the current in the phosphor layer is consistent with the applied anode-cathode current, and the current continuity is achieved. According to solid state physics, the accumulated charges in the phosphor layer should largely be trapped in the localized levels in the energy gap of the phosphor material, becoming space charges in the phosphor layer.[9]

In the low voltage cathodoluminescence, three typical voltages are involved, having the following relation, $V_0 = V_{acc} + V_{phosphor}$, where V_0 is the applied anode-cathode voltage, V_{acc} is the electron-accelerating voltage between the cathode and the phosphor layer, and $V_{phosphor}$ is the voltage drop on the phosphor layer. Assuming the exciting area of incident electrons on the phosphor layer is a unit, then the input power in cathodoluminescence should be $P_{input} = V_0 \cdot j$ and the exciting power in the phosphor layer should be $P_{excite} = V_{\text{acc}} \cdot j$, where j is the applied current density in low voltage cathodoluminescence. According to the related literature, the average penetration depth $\overline{X}$ of an incident electron in the phosphor layer is determined by the equation, $-\frac{dE_x}{dx} = \frac{2\pi NZe^4}{E_x} In \frac{E_x}{E_i}$, where E_x is the kinetic energy of incident electron for depth x.[10, 11] After simplification and calculation, $\overline{X} = a \cdot E_0{}^2$, where E_0 is the initial kinetic energy of the electron, and a is a constant depending on the phosphor material. In the low voltage cathodoluminescence, after the accelerating process from the cathode to the phosphor layer, the initial kinetic energy of an electron exciting the phosphor layer should be $E_0 = V_{acc} \cdot e$, where e is the electron charge. For the large amount of incident electrons being scattered randomly in the phosphor layer, the average penetration depth should be given to be,

$$\overline{X} = a \cdot E_0{}^2 = a \cdot V_{\text{acc}}{}^2 \cdot e^2 \qquad (1)$$

Assume the phosphor layer in low voltage cathodoluminescence is located at x=0 to x_d, x_d is the thickness of the phosphor layer, as shown in Fig. 1. Owing to the randomicity in the penetration depth distributions of incident electrons with an average $\overline{X}$, the incident current density $J_{incident}(x)$, which is composed of incident electrons before the kinetic energy is exhausted in the phosphor layer, should decrease gradually from j to 0 around $x = \overline{X}$ with the increasing of x (see Fig. 1), and the decreasing trend of $J_{\text{incident}}(x)$ should be proportionally consistent with that of penetration depth distributions. Consequently, the integration of $J_{incident}(x)$ should be given to be,

$$\int_0^{x_d} J_{incident}(x) \cdot dx = \overline{X} \cdot j \qquad (2)$$

In stable case, the continuity of current density should be achieved in the phosphor layer. Consequently, with the decreasing of $J_{incident}(x)$, another current density, i.e., the electric field-promoted intrinsic current density, should be increasing to achieve the continuity of current density, having the relation, $j = J_{incident}(x) + J_{promote}(x)$, where $J_{promote}(x)$ is the electric field-promoted intrinsic current density in the phosphor layer. Based on formula (2), the integration of $J_{promote}(x)$ is obtained to be,

$$\int_0^{x_d} J_{promote}(x) \cdot dx = \left(x_d - \overline{X}\right) \cdot j \tag{3}$$

The electric field $E(x)$ in the phosphor layer is determined by the density distribution $Q(x)$ of space charges in the phosphor layer, having the relation, $E(x) = \frac{1}{\varepsilon_0 \varepsilon_r} \int_0^x Q(x) \cdot dx$, where ε_0 and ε_r are the vacuum dielectric constant and the relative dielectric constant of phosphor material, respectively. Meanwhile, the electric field $E(x)$ can be obtained by the electric field-promoted intrinsic current density $J_{promote}(x)$ and the conductivity $\sigma(x)$ of the phosphor material, having the following relation,

$$E(x) = \frac{1}{\sigma(x)} \cdot j_{promote}(x) \tag{4}$$

In principle, with the incident electrons exciting the phosphor layer, the conductivity $\sigma(x)$ of the phosphor material should be improved in a certain extent owing to the generation of electron-hole (e-h) pairs.[12] Neglecting the variability in $\sigma(x)$, taking into account of formula (3), the voltage drop $V_{phosphor}$ on the phosphor layer should be given to be,

$$\begin{aligned} V_{phosphor} &= \int_0^{x_d} \frac{1}{\sigma(x)} \cdot J_{promote}(x) \cdot dx \\ &= \frac{1}{\sigma} \cdot \left(x_d - \overline{X}\right) \cdot j \end{aligned} \tag{5}$$

In the low voltage cathodoluminescence, e-h pairs would be generated (with generation rate g_{e-h}) in the phosphor layer owing to the energy input of incident electrons. The generated e-h pairs may transfer their energy (with energy transfer probability η_{et}) to excite the ground-state activators or be recombined by sending phonons. The excited activators would transit to the ground states radiatively (with radiative transition probability η_{rad}) or lose their energy in the host lattice.[13] The output power, i.e., the output luminance flux in cathodoluminescence should be given to be,

$$P_{output} = g_{e-h} \cdot \eta_{et} \cdot \eta_{rad} \cdot hv \qquad (6)$$

Where hv is the energy of a photon emitted in cathodoluminescence.

3.2 Saturation behavior of exciting power owing to the low conductivity of the phosphor material in low voltage cathodoluminescence

Based on formula (1), the voltage difference $V_0 - V_{\mathrm{acc}}$, as the function of $\overline{X}$, is depicted with a parabola, as shown in Fig. 2. Meanwhile, the voltage drop $V_{phosphor}$, linearly dependent of $\overline{X}$ in formula (5), is depicted to present a different changing tendency. In view of the relevance in the three typical voltages ($V_0 = V_{acc} + V_{phosphor}$), the conditionality in $\overline{X}(j)$ is visually presented in Fig. 2. It could be seen that for a fixed anode-cathode voltage V_0, the average penetration depth $\overline{X}(j)$ would decrease from its upper bound $\mathrm{a} \cdot V_0{}^2 \cdot e^2$ to 0 with the increasing of j from 0 to its upper bound $\frac{V_0 \cdot \sigma}{\mathrm{x}_d}$ (θ increasing from 0 to $\mathrm{arc}tg\frac{V_0}{x_d}$). On the other hand, it could be seen that the average penetration depth $\overline{X}(j)$ would be larger for an improved conductivity σ of the phosphor material. When the conductivity σ is rather higher, $\overline{X}(j)$ would tend to be a constant $\mathrm{a} \cdot V_0{}^2 \cdot e^2$, independent of j. As visualized in Fig. 2.

Based on formulas (1), (5) and the quantitative relations, $V_0 = V_{acc} + V_{phosphor}$, $P_{excite} = V_{\mathrm{acc}} \cdot j$, the exciting power P_{excite} in low voltage cathodoluminescence could be calculated to be,

$$P_{excite} = \frac{\sigma - \sqrt{\sigma^2 + 4a \cdot e^2 (x_d \cdot j - V_0 \cdot \sigma) \cdot j}}{2a \cdot e^2} \qquad (7)$$

The exciting power P_{excite}, together with the input power P_{input} in cathodoluminescence, is depicted as the function of j in Fig. 3. It could be seen that for a fixed anode-cathode voltage V_0, the exciting power P_{excite} increases near-linearly at first with the increasing of j, and then it tends to saturate until it reaches its peak value, and then begins to decrease with the increasing of j. It could be calculated that the exciting power reaches its peak value $P_{excite}^{\max} = \frac{1-\sqrt{1-a\cdot e^2\cdot {V_0}^2/x_d}}{2a\cdot e^2}\cdot\sigma$ when $j=\frac{V_0}{2x_d}\cdot\sigma$, as indicated in Fig. 3. The specific current density threshold $j_{th}=\frac{V_0}{2x_d}\cdot\sigma$ for the peak exciting power is a critical parameter, by comparing the applied current density j with this parameter, the nonlinear degree of exciting power could be determined. It should be noted that in the operation in cathodoluminescence, the applied current density j should be relatively smaller than $j_{th}=\frac{V_0}{2x_d}\cdot\sigma$. Consequently, the saturation behavior in exciting power with the increasing of j is what may happen actually.

It is apparent that when the applied anode-cathode voltage V_0 is higher and the applied current density j is smaller, then the threshold $j_{th}=\frac{V_0}{2x_d}\cdot\sigma$ for the peak exciting power would be larger and j may be much smaller relative to j_{th}, and the saturation behavior in exciting power would not emerge (see Fig. 3). This is the true case in cathode ray tube. On the contrary, when V_0 is lower and j is larger, then the threshold $j_{th}=\frac{V_0}{2x_d}\cdot\sigma$ would be smaller and j would not be relatively too small in contrast to j_{th}, and the saturation behavior in exciting power would be exhibited (see Fig. 3). This is the true case in field emission display. The saturation behavior in exciting power P_{excite} means the loss of input power P_{input}, and the lost portion is the thermal consumption power originated in the low conductivity of the phosphor material.

We can see the current density threshold j_{th} and the peak exciting power $P_{excite}^{\max}$ are the key parameters determining the dynamic performance of exciting power $P_{excite}(j)$ in the low voltage cathodoluminescence. The larger these two parameters, the larger the variation ranges of j and P_{excite} that can be achieved in cathodoluminescence, as shown in Fig. 3. It should be noted that these two parameters are all proportional to the conductivity σ of the phosphor material. For the typical phosphor materials applied in cathodoluminescence, a much lower conductivity is dominated in the magnitude of 10^{-6}~10^{-9} S/cm, therefore, they should be classified to be insulators.[13] Zinc oxide (ZnO) is a green-emitting phosphor material, having higher conductivity, while ZnO is unfit for the full-color display in the low voltage cathodoluminescence.[14] If the conductivity σ of the phosphor material were to be significantly improved, then the key parameters, j_{th} and $P_{excite}^{\max}$, may be elevated simultaneously to rather larger values, even if the applied anode-cathode voltage V_0 is kept at a low level in cathodoluminescence. In this case, the linear region of $P_{excite}(j)$ is enlarged and the deviation between P_{excite} and P_{input} would tend to vanish owing to the enlarged difference between j and j_{th} (j is much smaller relative to j_{th}). This means the saturation behavior in exciting power P_{excite} may be restrained, i.e., the loss in P_{input} may be saved by improving the conductivity σ of the phosphor material. It should be noted that the phosphor layer is not appropriate to be made excessively thin to improve the key parameters (j_{th} and $P_{excite}^{\max}$), because the thickness x_d of the phosphor layer should be sufficiently larger than $\mathrm{a} \cdot V_0{}^2 \cdot e^2$, which is the upper limit of average penetration depth $\overline{X}$ of incident electrons in the phosphor layer. Consequently, to improve the conductivity σ of the phosphor material is the ultimate method to restrain the saturation behavior in exciting power in the low voltage cathodoluminescence.

3.3 Degraded performance owing to the low conductivity of the phosphor material in low voltage cathodoluminescence

In the low voltage cathodoluminescence, e-h pairs would be generated in the phosphor layer owing to the energy input of incident electrons. The generated e-h pairs would be recombined at the recombination centers or transfer their energy to excite the ground-state activators. The excited activators would then transit to the ground states radiatively or lose their energy in the host lattice.[13]

In the excitation of incident electrons in the phosphor layer, the average energy required to generate an e-h pair is $\beta_g \cdot E_g$, where E_g is the energy gap of the phosphor material and β_g ranges from 2.7 to 5, depending on the phosphor material.[13] Consequently, the generation rate of e-h pairs in the phosphor layer should be given to be,

$$g_{e-h} = P_{excite} / \left(\beta_g \cdot E_g\right) \tag{8}$$

Since g_{e-h} is proportional to the exciting power P_{excite}, the saturation behavior of P_{excite} would directly lead to the saturation behavior of g_{e-h} in the low voltage cathodoluminescence.

In the low voltage cathodoluminescence, the average penetration depth $\overline{X}$ of incident electrons may be seen to be the thickness of excitation region beneath the surface of the phosphor layer. Since P_{excite} increases and $\overline{X}$ decreases with the increasing of j, the excitation intensity, i.e., the excitation power allocated in unit volume in the excitation region, given in equation (9), would increase swiftly with the increasing of j.

$$I_{excite} = \frac{P_{excite}}{\overline{X}} \tag{9}$$

With the increasing of I_{excite}, ground-state depletion of activators would emerge, i.e., the majority of the activators in the excitation region would be in the excited states. In this case the concentration of generated e-h pairs is very large while the concentration of ground-state activators is very low in the excitation region. As a result, the energy transfer probability η_{et}

of an e-h pair exciting a ground-state activator is decreased. If the conductivity σ of the phosphor material were to be significantly improved, then $\overline{X}$ would tend to be a constant $a \cdot {V_0}^2 \cdot e^2$ and P_{excite} would tend to increase linearly with j. As a result, I_{excite} would tend to increase linearly with j. This means the swift increasing of I_{excite} should be restrained, causing a slower decrease in the energy transfer probability η_{et}.

As aforementioned, the saturation behavior of exciting power means the loss of input power, and the lost portion is the thermal consumption power originated in the low conductivity of phosphor material, resulting in an increased temperature of the phosphor layer. Meanwhile, the decrease in energy transfer probability η_{et} of e-h pairs means a loss of exciting power, resulting in a further increased temperature of the phosphor layer. As a result, the radiative transition probability η_{rad} of excited activators would be decreased owing to the increase of temperature. Similarly, the nonradiative transition of excited activators is another loss of exciting power, resulting in a further increase of temperature, and hence a further decrease of η_{rad}. It is apparent that if the conductivity σ of the phosphor material were to be significantly improved, these effects causing the temperature increasing of phosphor layer would be eliminated in a certain extent. As a result, the decrease in η_{rad} would be restrained.

It has been pointed out that the average energy required to generate an e-h pair is $\beta_g \cdot E_g$ in cathodoluminescence, and β_g ranges from 2.7 to 5. This means the maximum efficiency of incident electrons generating e-h pairs is $1/2.7 \approx 37\%$. It should be noted that the efficiency factor β_g is an intrinsic parameter of the phosphor material related to the thermal consumption and increasing of temperature, be a constant in cathodoluminescence.

So far, the physical mechanisms in the conductivity-dependent performance in low voltage cathodoluminescence have been revealed. First, the exciting power P_{excite} tends to saturate with the increasing of j, causing the saturation behavior of e-h generation rate

g_{e-h}. Second, for a generated e-h pair, the energy transfer probability η_{et} to excite a ground-state activator would decrease with the increasing of j. Third, for an excited activator, the radiative transition probability η_{rad} would decrease with the increasing of j. In view of these key factors, the saturation behavior in the output luminance flux, given in formula (6), should be the inevitable result. To restrain the saturation behavior, so as to improve the performance in low voltage cathodoluminescence, the conductivity of the phosphor material were to be improved.

3.4 Improving the conductivity of phosphor material to improve the performance in low voltage cathodoluminescence

In our experiments, taking Y_2O_3:Eu^{3+} as the typical phosphor material in cathodoluminescence, a series of experiments have been carried out. According to the electric percolation theory of composite conductive materials, a suitable conductive material, as a separate component, may be introduced into the phosphor to improve the conductivity of the phosphor material.[6, 15–19] Among the traditional candidate materials, In_2O_3, which is a transparent wide band gap semiconductor (Eg=3.6eV), has been proven to be an effective conductive component due to the high conductivity it presents ($\sigma = \sim 10^3\ S/cm$), the related characterizations of In_2O_3-introduced phosphor materials have been given in our previous research work and reported in literatures.[6, 20, 21] Our previous reseach work indicated that the distribution of In_2O_3 is not a uniform layer but is island-like condensates distributed on the phosphor surfaces.[21] The formation reason of In_2O_3 island-like condensates lies in that In_2O_3 material has a relative higher surface energy, so that In_2O_3 component may be drifted and shrunken to islands on the phosphor surface in the annealing process. According to the percolation theory of composite conductive materials, the conductive component in composite materials forms conductive channels and the conductive channels form three-dimensional network structures in composite materials, so that the electrical conductivity of composite materials is increased.[21] The results in resistivity measurements confirmed the conductivity increasing with In_2O_3 content (0%, 1%, 2%, 3%, 4%, 5%) in Y_2O_3:Eu^{3+} phosphor material, as shown in Fig. 4.

It should be noted that the introduced conductive component, as heterogeneous impurity, would cause negative effect on the efficiency of the phosphor material, even though a higher conductivity is achieved. It can be expected that when j is lower, so that the saturation behavior in P_{excite} would not emerge, then the degraded efficiency of the phosphor material due to the introduction of conductive component should be the dominant effect in the low voltage cathodoluminescence. Conversely, when j is larger, the improved conductivity of the phosphor material due to the introduction of conductive component should be the dominant effect improving the performance in low voltage cathodoluminescence. These predictions have been confirmed in the experiments presented in Fig. 5, in which, based on the discrete experimental data, the continuous curves indicating the cathodoluminescent intensities have been depicted for Y_2O_3:Eu^{3+} phosphor with different In_2O_3 contents (0%, 1%, 2%, 3%, 4%). It can be seen that the relative cathodoluminescent intensities of Y_2O_3:Eu^{3+} phosphor with different In_2O_3 contents are changed with the increasing of j. The typical emission spectra in low voltage cathodoluminescence have been given in Fig. 6. The linear region of the continuous curve in Fig. 5 is extended with the conductivity increasing of the phosphor material, indicating the efficiency in cathodoluminescence remains to be a constant for a larger variation range of j. Consequently, the saturation behavior was effectively restrained and the performance in low voltage cathodoluminescence was sufficiently improved. Meanwhile, it is apparent that the proper amount of conductive component to be introduced in the phosphor material is determined by the specific variation range of the applied current density j in the low voltage cathodoluminescence.

4. Conclusions

Based on the comprehensive investigations, the physical mechanisms in the conductivity-dependent performance in low voltage cathodoluminescence have been objectively revealed. With the increasing of applied current density in the low voltage cathodoluminescence, the e-h generation rate would tend to saturate. Meanwhile, the energy transfer probability from e-h pairs to the ground-state activators would be decreased, and the radiative transition probability of excited activators would be decreased. To restrain the saturation behavior in low voltage cathodoluminescence, the ultimate method is to improve the conductivity of the phosphor material. By introducing In_2O_3 conductive component into the typical Y_2O_3:Eu^{3+} phosphor material, improved cathodoluminescence was effectively achieved.

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Figures:

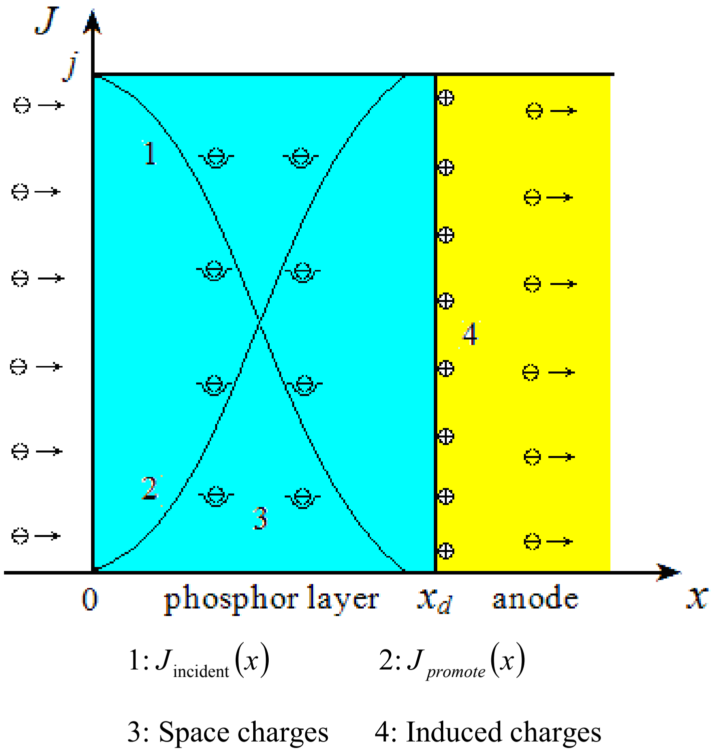

Fig. 1 Schematic diagram of current density distributions and charge accumulations.

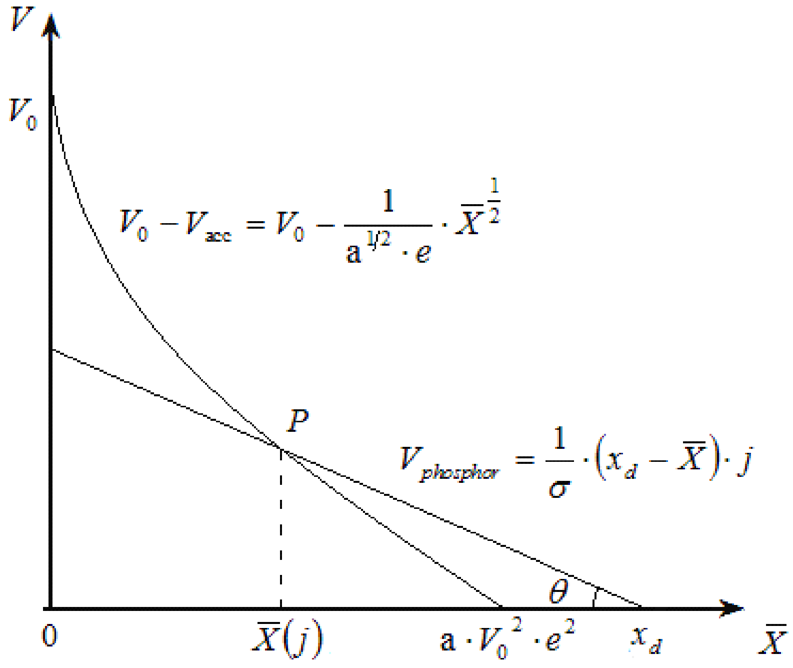

Fig. 2 Voltage difference $V_0 - V_{acc}$ and voltage drop $V_{phosphor}$ as the functions of $\overline{X}$.

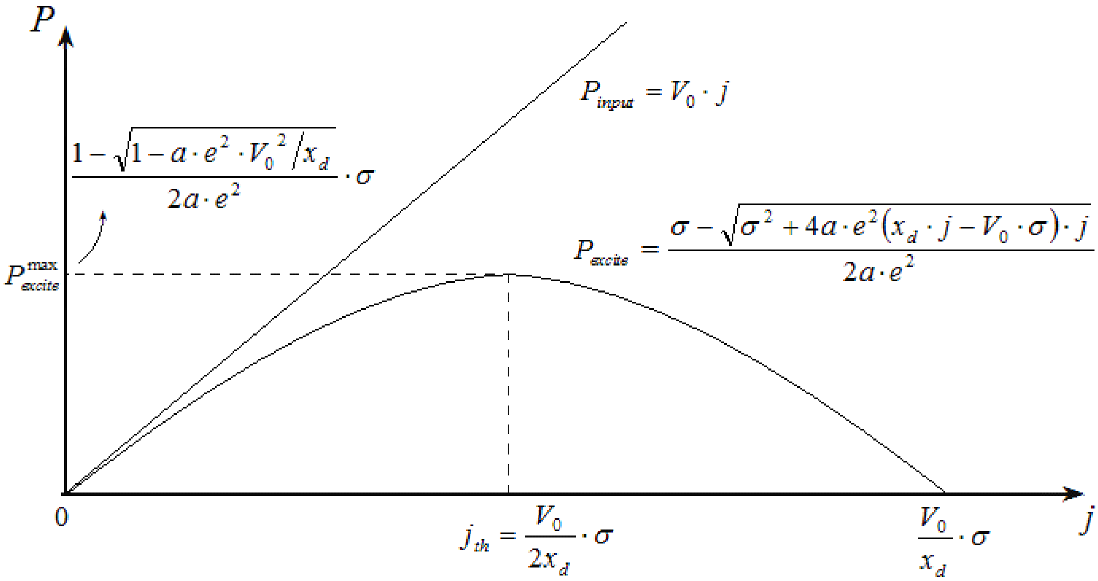

Fig. 3 Exciting power P_{excite} and input power P_{input} as the functions of j.

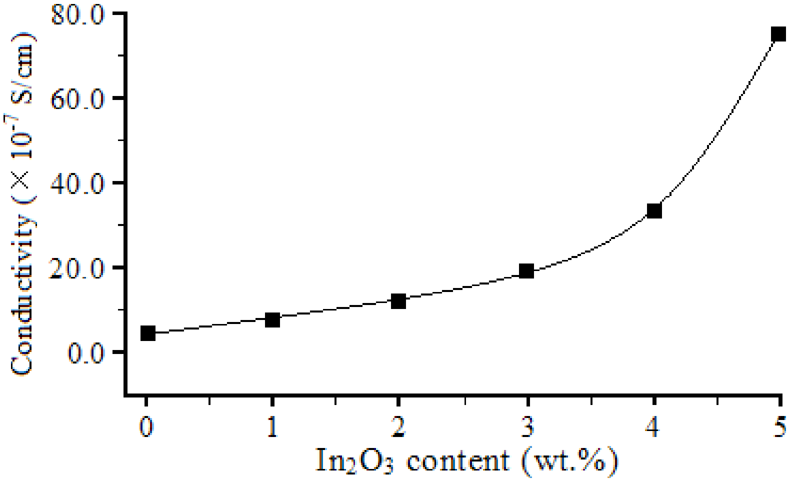

Fig. 4 Conductivity increasing with In_2O_3 content in Y_2O_3:Eu^{3+} phosphor material.

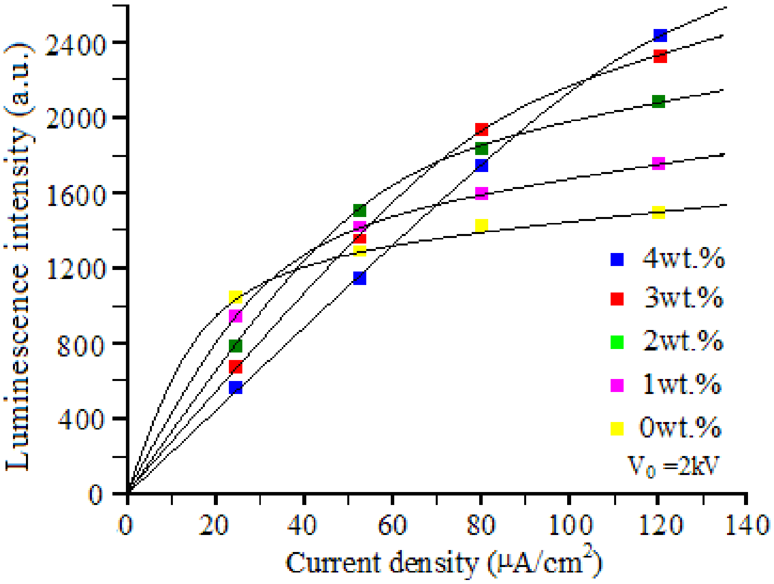

Fig. 5 Cathodoluminescence intensity-current density relations of $Y_2O_3:Eu^{3+}$ phosphor with different In_2O_3 contents of 0wt.%, 1wt.%, 2wt.%, 3wt.% and 4wt.% at a fixed voltage of 2 kV.

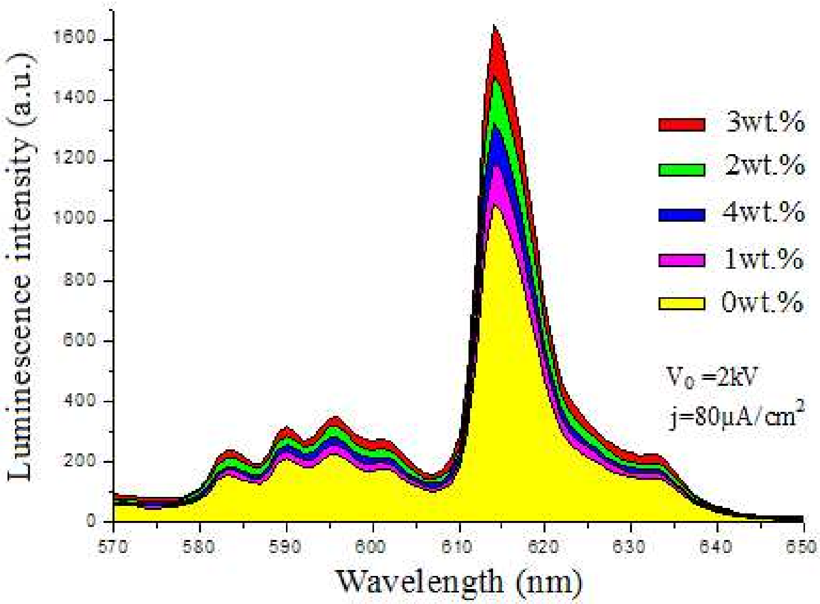

Fig. 6 Emission spectra of $Y_2O_3:Eu^{3+}$ phosphor with different In_2O_3 contents of 0wt.%, 1wt.%, 2wt.%, 3wt.% and 4wt.% at a fixed voltage of 2kV and a fixed current density of 80μA/cm^2.