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Introduction of cooperating conductive components into the phosphor to improve the low voltage cathodoluminescence

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ABSTRACT

In order to improve the electric conductivity of $Y_2O_3:Eu^{3+}$ phosphor with the least amount of conductive component so as to maximize the improvement in low voltage cathodoluminescence, In_2O_3 and Cu nanowires (NWs) were simultaneously introduced to form Cu NWs/ In_2O_3 -attached $Y_2O_3:Eu^{3+}$ phosphor. In_2O_3 and Cu NWs play different roles in the formation of electrically conductive network, i.e., Cu NWs are suitable as conductive channels for charge transmission due to their one-dimensional morphology with large slenderness ratios, while the island-like In_2O_3 condensates form local conductive contacts joining the adjacent Cu NWs. Meanwhile, In_2O_3 forms attachment between Cu NWs and the phosphor. Owing to the cooperating effects between Cu NWs/ In_2O_3 conductive components in the phosphor, the efficiency in low voltage cathodoluminescence was significantly improved.

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1. Introduction

Field emission display (FED) has been developed as a next-generation flat panel display; it combines the high image quality of cathode ray tube (CRT) with the thin-thickness/light-weight properties of flat panel display [1–4]. Specifically, FED should be operated at a low voltage (< 4 kV); hence a large current density (10 – $100 \mu A/cm^2$) is required to guarantee a high power in luminance output. However, cathodoluminescence operated at low voltage and large current density typically exhibits luminance saturation behavior. The saturation behavior in low voltage cathodoluminescence means decrease in efficiency with the increase of current density. This is one of the most serious impediments hindering the development in FED. In our previous research work, we have revealed in several aspects that the saturation behavior in low voltage cathodoluminescence is originated from the insulation of phosphor. The correlations between the insulation of the phosphor and the saturation behavior in cathodoluminescence have also been reported in literatures [3,5]. In this paper, two kinds of conductive components with different morphologies were introduced into $Y_2O_3:Eu^{3+}$ phosphor at the least amount. Due to the cooperating effects between the

conductive components, the electrical conductivity of $Y_2O_3:Eu^{3+}$ phosphor was significantly increased and the low voltage cathodoluminescence was effectively improved.

2. Experimental section

2.1. Synthesis of $Y_2O_3:Eu^{3+}$ phosphor

$Y_2O_3:Eu^{3+}$ ($(Y_{0.97}Eu_{0.03})_2O_3$) powder phosphor was prepared by glycine–nitrate solution combustion synthesis [6]. In brief, analytical grade 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 5:3) to form the precursor solution. The precursor solution was concentrated by heating at $260^\circ C$ until the excess free water evaporated and spontaneous combustion occurred. After annealing at $800^\circ C$ for 1 h, $Y_2O_3:Eu^{3+}$ powder phosphor with an average size in micron scale was formed.

2.2. Preparation of In_2O_3 -attached, Cu NWs/ In_2O_3 -attached and Cu NW-mixed $Y_2O_3:Eu^{3+}$ phosphor

$Y_2O_3:Eu^{3+}$ phosphor was attached to In_2O_3 by hydrolysis of indium isopropoxide ($In[OCH(CH_3)_2]_3$) in sol–gel process [3]. $Y_2O_3:Eu^{3+}$ powder phosphor, H_2O for hydrolysis and isopropyl alcohol were added into the alkoxide solution of In. During churning of these reactants at $80^\circ C$, hydrolysis and condensation

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occurred. After drying the solution and heating at 300 °C for 3 h, In_2O_3 -attached $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ phosphor was formed.

In the preparation procedure of In_2O_3 -attached $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ phosphor, a certain amount of ethanol solution of Cu NWs (around 50 nm in diameter and around 2.0 μm in length) which had been sonicated for 2 h was added into the alkoxide solution of In; then Cu NWs/ In_2O_3 -attached $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ phosphor was obtained in the end. Cu NWs were purchased from XianFeng Nano-material Technology Co., Ltd.

Ethanol solution of Cu NWs which had been sonicated for 2 h was added with $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ phosphor; after a further sonication process of 2 h, the solution was dried and heated at 500 °C for 3 h; Cu NW-mixed $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ phosphor was thus obtained.

2.3. Characterization

In order to determine the resistivity of In_2O_3 -attached, Cu NWs/ In_2O_3 -attached and Cu NW-mixed $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ phosphor, each kind of phosphor (0.5 g) was made into a cylinder with a diameter $2r=13$ mm and thickness $d \approx 2$ mm under a pressure of 1.5×10^2 MPa; the electrodes were made by Al deposition on both sides of each cylinder. The resistance for each cylinder was measured by a 769YP-24B megohmmeter. The resistivity was calculated by the following equation:

$$\rho = \frac{\pi r^2}{d} R$$

The crystalline phases of the phosphor were identified by a Rigaku-D/max 2500 X-ray diffractometer using Cu K α radiation ($\lambda=0.15405$ nm). The morphology of the phosphor was characterized using a KYKY-1000B scanning electron microscope. The CL measurements were carried out in an ultra-high-vacuum chamber ($< 10^{-8}$ Torr), where the phosphor was excited by an electron beam in a range of 2.5–5 kV with different filament currents, and the spectra were recorded using an F-7000 spectrophotometer.

3. Results and discussion

3.1. Saturation behavior in low voltage cathodoluminescence due to insulation of the phosphor

For the cathodoluminescence in field emission display, the phosphor layer on the anode surface is excited by the energy input of electron beam emitted from the cathode. Under the electron beam excitation, electron–hole (e–h) pairs would be generated in the phosphor layer. The generated e–h pairs may be recombined at the recombination centers or transfer their energy to excite the activators. The excited activators would then transit to the ground states radiatively or lose their energy in the host lattice.

Assuming that the exciting area of electron beam on the phosphor layer is a unit, the input power should be $P_{input}=V_0j$, where V_0 is the anode–cathode voltage and j is the current density. However, the exciting power of electron beam on the phosphor layer is given by

$$P_{excite} = (V_0 - \rho dj)j \\ = V_0j - \rho dj^2$$

where ρdj is the voltage drop on the phosphor layer, ρ is the resistivity of the phosphor material and d is the thickness of the phosphor layer. The exciting power P_{excite} , as a function of current density j , should be depicted as a parabola, as shown in Fig. 1. With the increase of current density j approaching the current threshold $j_{th} = V_0/2\rho d$, a saturation behavior in exciting power would be exhibited. The saturation behavior of exciting power

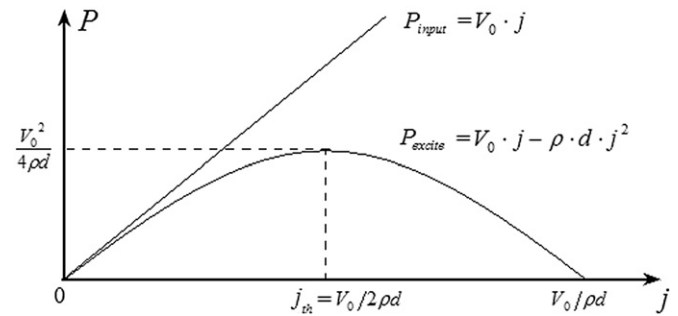


Fig. 1. Exciting power P_{excite} and input power P_{input} as functions of current density j .

results in a series of effects in the phosphor layer. The generation rate g_{num} of e–h pairs in the phosphor layer, the energy transfer probability η_{et} of an e–h pair exciting an activator and the radiative transition probability η_{rad} of an excited activator would all be decreased due to the saturation behavior of exciting power with the increase of current density j [7–9]. Consequently, the output luminance flux in cathodoluminescence, which is given by

$$P_{output} = g_{num}\eta_{et}\eta_{rad}h\nu$$

would tend to saturate with the increase of current density j .

It is apparent that when the applied voltage V_0 is higher and the current density j is smaller, then the current density threshold $j_{th} = V_0/2\rho d$ for the peak exciting power would be larger and j may be much smaller than j_{th} , and the saturation behavior in exciting power would not emerge (see Fig. 1). This is the true case in cathode ray tube (CRT). On the contrary, when the applied voltage V_0 is lower and the current density j is larger, the threshold $j_{th} = V_0/2\rho d$ would be smaller and j would not be relatively too small in contrast to j_{th} , and a saturation behavior in exciting power would be exhibited. As a result, the saturation behavior in cathodoluminescence would be apparent. This is the actual case in field emission display (FED).

For ordinary phosphors used in cathodoluminescence, the phosphors have a much higher resistivity with a magnitude of 10^6 – 10^9 Ω m. Consequently, they should be classified as insulators. If the resistivity ρ of the phosphor material were to be significantly lowered, then the current threshold $j_{th} = V_0/2\rho d$ for the peak exciting power may be elevated to a larger value, even if the applied voltage V_0 is kept at a low level. It should be noted that it is not appropriate for the phosphor layer to be made too thin; this means the thickness d is limited and should not be very small. Consequently, to lower the resistivity of the phosphor material is the ultimate method to restrain the saturation behavior in exciting power, so as to restrain the saturation behavior in low voltage cathodoluminescence.

3.2. Marginal improvement in cathodoluminescence achieved by introduction of In_2O_3 conductive component in $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ phosphor

In our research work, taking $\text{Y}_2\text{O}_3:\text{Eu}^{3+}$ as the typical phosphor in cathodoluminescence, a series of experimental investigations have been carried out. In principle, to lower the resistivity of the phosphor so as to improve the low voltage performance in cathodoluminescence, a suitable conductive material, as a separate component, may be introduced into the phosphor [3,10–13]. Among the traditional candidate materials, In_2O_3 , which is a transparent wide band gap semiconductor ($E_g=3.6$ eV), has been proven to be a kind of effective conductive component due to its high electrical conductivity ($\rho \sim 10^{-3}$ Ω m) and its favorable compatibility in preparation and attachment with the phosphor.

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