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Reliability of AC thick-film electroluminescent lamps

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Abstract. The reliability of AC thick-film EL devices has been studied. The AC thick-film EL devices were fabricated by Novatech Inc. using the industrial print screen technology. The analysis of reasons for failure has been proposed. The dependence of EL lamp parameters on physical properties of the device EL layers was found. Our analysis of the breakdown spot showed that improvement of reliability can be reached using the additional dielectric layer between the phosphor layer and transparent electrode, high concentration of phosphor powder 70 % and binder 30 %, balanced resistance between the electric circuit and EL lamp. The thickness of the phosphor layer was equal to $H = (1 + \sqrt{3}/2)D$ (hexagonal packing), where D is the mean diameter of phosphor particles. The reliability dependence of EL lamp on a water adsorption property of packaging material was revealed.

Keywords: electroluminescence, thick film, reliability, ZnS, lifetime.

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1. Introduction

The electroluminescent (EL) AC thick films on flexible substrates are widely used as light sources in indicator devices, back light for liquid crystal display panels and for advertisement [1]. So, reliability and the operation lifetime are main parameters in applications of AC EL devices. Optical and electrical properties of AC EL lamp have been studied by numerous authors but the factors providing high reliability are not yet clear [2, 3]. The enhanced reliability and lifetime are the main task in developing new EL devices. First of all, it is required to ascertain the factors causing a damage of EL devices and ways to avoid them.

The purpose of this work is to test the lifetime of EL devices, analyze failure mechanisms and to find how the EL lamp parameters depend on physical properties of the EL device layers.

2. Experimental

Tested in this work were the samples manufactured EL-Korea Corp. and Novatech INC. using the industrial print screen technology based on Sylvania and Durel phosphors. The tests were carried out by the following way. The lots of samples fabricated in the same conditions were separated by two sets. The first set was tested in normal conditions for 2500 hours in every sample set. The second set was put into a humidity chamber at the temperature 60 °C and 90 % humidity. All the samples were applied by the voltage 100 V and frequency 400 Hz. The failure under observation was complete or partially loss of brightness. In this case, the samples were removed from the further testing.

Analyzing the mechanism of these failures, we considered two kinds of the latter: “fast” and “slow” ones. As a “fast” failure, we imply the sudden and complete loss of the brightness. The term “slow” failures means a slow drop of the brightness caused by aging or degradation processes. When the brightness of a device became lower than 50 % as compared to the initial one, we called such a defected sample as “slow” failure. The measurement of quantity relation of the spoilt samples (η_t) was carried out for a set of the tested samples at normal conditions every 25 hours (Δt = 25 hours) and for the set of the samples at high humidity every 2 hours (Δt = 2 hours). The calculation was made by the following way. Let us assume that N samples were taken for testing. After 25 hours, n_1 samples were defected. On the $\eta(t)$ curve the first point $\eta_1 = n_1 / N$ at the $t_1 =$ 25 hours has been found. The next is $\eta_2 = n_2 / (N - n_1)$ point at the $t_2 = 50$ hours. The unit n_2 is the number of defected samples for the time interval (25 hours) and $(N - n_1)$ is the initial number of samples at t_1. Thus, at any moment t the value η_t is the number of defected samples for 25 hours (n_i) divided by the number of taken samples (N_i) tested at the moment $t = 25$ hours.

A Luminance Colorimeter BM-7 (TOPCON, Tokyo Optical Co., LTD, Japan) and SpectraColorimeter (PR-650. Photoresearch, Inc., USA) were used to determine the brightness of EL lamps. Using the humidity chambers (DF-961HC-MJ. Dury Scientific Co., Korea) and AC-7602HA-1X82 (Blue M Electric, USA), the reliability test was performed.

3. Results and discussion

Fig. 1 shows the faulty EL devices for two lots of samples made using fluoresine and cyanoresine as a binder. Here, η_t is a ratio of the spoilt samples to the taken quantity of samples during the test time Δt. For the first 400 hours, practically all the failures happened. Moreover, the maximum of failure rate over the interval 150-250 hours was observed. As a rule, the "sudden" failures occurred within this time interval. The time interval (from 800 to 2000 hours) is characterized by a very small quantity of the faulty samples. Therefore, the operation capacity of the EL devices becomes stable. The growth of the failure rate within 2200 hours is a result of the fast decrease in the brightness (below half lifetime), the so-called degradation processes. Therefore, after 2000-2100 hours of operation the EL devices must be replaced, because the probability of their failure increases very fast. The failure curve at high humidity looks like that for normal conditions but is very strongly time compressed. The maximum of the failure rate is observed for 30 hours of operation. All the failures before 30 hours may be considered as the "fast" ones.

The "sudden" failures are a major cause for the break of EL devices during the first hundred hours of applying the voltage. Our analysis of the "sudden" failures allows to separate two kinds of them. The first one concerns the "sudden" failures caused by electrical breakdown of layers. And the second one is characterized by a loss of electrical connection. Approximate evaluation of the probability for every kind of failures showed that the probability to lose the electrical contact is 15 to 20 times less than the probability of failures caused by the electrical breakdown of layers. Consequently, the electrical breakdown of layers is the main process responsible for restriction of operation life for these EL devices.

Since, the electric field inside the phosphor layer is very high (~10^5 V/cm), it can be assumed that the breakdown spots appear in the places where disturbances of uniformity are located. Thus, the disturbance of homogeneity in the layer density can happen in layers fabricated using the print screen technology. This kind of breakdown spots is called as "pinholes". Usually, pinholes are observed in places where the thickness of dielectric layer is very thin. It is occasionally formed by phosphor grains as a result of burning off the dielectric layer. The electric field in dielectric layer is higher than the average field in the EL one, which forms suitable conditions for electrical breakdown. Besides, there is a possibility for penetration of the back electrode material into EL layer during the printing process. It can be a reason for breakdown, too. Shown in Fig. 2 is the photo of a breakdown voltage spot.

The crater with the outer diameter of 30-50 μm and the inner one of 5-10 μm was observed on the back graphite electrode. On the front transparent electrode (ITO) of the EL device, separation of the phosphor layer was observed. This separation looked as a crater with the external diameter of 200 μm. There is the black spot of 20 μm diameter in the center. Here, one can see a drowned part of the electrical breakdown channel in the EL structure (Fig. 3).

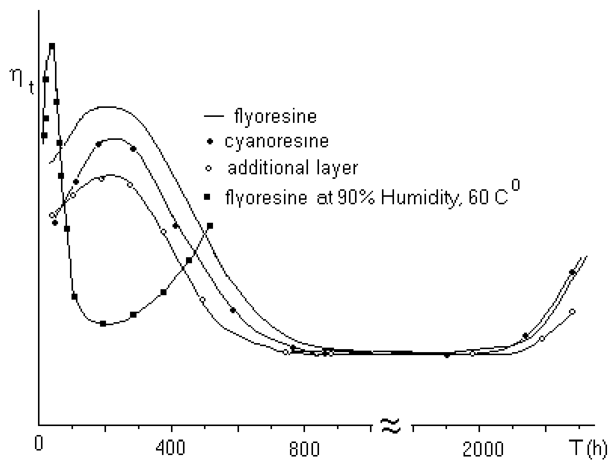

Fig. 1. Distribution of the failures in time.

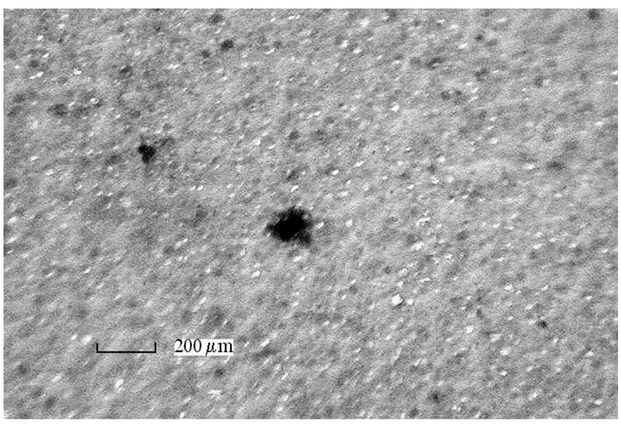

Fig. 2. Photo of a breakdown voltage spot.

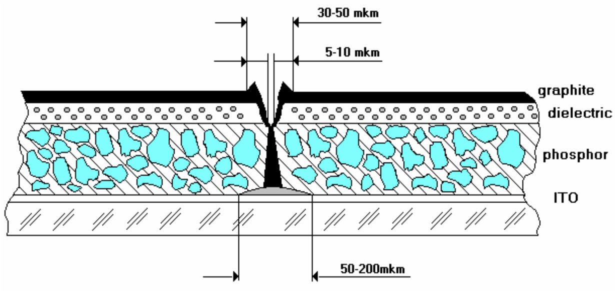

Fig. 3. Section of the electrical breakdown channel.

Oscillographic testing the breakdown process showed that the current 20 mA is achieved during the time of breakdown. Electrical breakdown is observed due to a charge transfer from the back electrode to the front one through the system layer. The value of this current depends on ohmic resistance of electrodes and on the reaction rates at the electrode-polymer interfaces. It should be noticed that at high current the almost full voltage of electrical source on the breakdown spot is applied, i.e. a phosphor layer or rather ZnS grains contain a free charge, but the dielectric layer (insulator) does not contain a free charge. Electrons possessing a high kinetic energy make the mechanical damage like a spherical figure of the “crater form” (Fig. 3). The electrical resistance of the breakdown spot was of the order of several kOhm, while the resistance in undamaged areas was higher than a few tens MOhm. Thus, to increase reliability of the EL device it is necessary to make a good electrical stability of the dielectric layer. Analyzing different constructions of EL lamps, we concluded that the increase in reliability can be reached by a few ways. First of all, it is necessary to select a balancing resistance between the electric circuit and EL lamp. Taking into account the resistance of EL lamp and basing on the minimum breakdown current (20 mA), it is easy to select a peak value of the balancing resistance. The value of the balancing resistance is defined from transient processes at a power up time of the current source when the EL lamp is short-circuited. The balancing resistance value should be defined by a minimum breakdown current. It is necessary to mark that using the balancing resistance makes sense for EL lamps with the small emitting areas, i.e. when the current is not less than 20 mA. This method can increase the reliability up to 20 %.

The trivial way for reaching the electrical stability of EL devices is an increase of the layer thickness in EL devices. But usually it leads to decrease in brightness. One of the ways is creation of an insulating layer (we call it as an additional dielectric layer) between the phosphor layer and the transparent front electrode. The additional dielectric layer not only increases a reliability of EL devices, but also considerably improves uniformity of the phosphor layer. To understand the voltage distribution in an EL cell with the additional dielectric layer, we consider the EL cell that contains two layers under applied electric field. The equivalent electric scheme of the cell can be represented as two capacitors connected in series (Fig. 4). The voltage of the source will be distributed between these capacitors in inverse proportion to their electrical capacitance [4].

$$V_{EL} / V_I = C_I / C_{EL} \,, \tag{1}$$

where V_{EL} is the voltage drop in the phosphor layer, V_I is the voltage drop in the additional dielectric layer, C_{EL} and C_I are their capacitances. The capacitance of a flat capacitor is expressed by the conventional formula

$$C \sim \varepsilon S / d \,, \tag{2}$$

where ε is the dielectric constant of two layers, S is the area of layers and d is the distance between layers. Using (2) we can write

$$V_{\mathbf{EL}} / V_{\mathbf{I}} = (\varepsilon_{\mathbf{I}}\, d_{\mathbf{EL}}) / (\varepsilon_{\mathbf{EL}}\, d_{\mathbf{I}}), \tag{3}$$

where ε_{EL} and ε_I are the dielectric constants of the phosphor layer and additional dielectric layer, respectively, and d_{EL}, d_I are their thicknesses. Because of the electric field in the capacitor is $E = V / d$, the expression (3) may be rewritten as

$$E_{EL} / E_I = \varepsilon_I / \varepsilon_{EL} \,. \tag{4}$$

The additional dielectric layer increases the common electrical stability of the EL device and, consequently, its reliability. However, in this case it is necessary to increase the applied voltage, because the voltage drop occurs in the additional dielectric layer

$$V_{\mathbf{I}} \sim 1/C_{\mathbf{I}} \sim d_{\mathbf{I}} / \varepsilon_{\mathbf{I}} \,. \tag{5}$$

The voltage drop (V_I) is increased with the thickness of the additional dielectric layer. Therefore, the voltage drop in the light-emitting layer ($V_{EL} = V_{source} - V_I$) at the same applied voltage is decreased, and it causes the decrease in brightness. So, it is necessary to use the additional dielectric layer materials with a high dielectric constant to decrease the applied voltage ($V_{\mathbf{I}}$). At equal thicknesses of phosphor and dielectric layers, the EL lamp brightness was measured in the dependence on the complex dielectric constant of the additional dielectric layer. In this case, due to high values of the dielectric constant in an additional polymer layer, the increase of the electric field was reached. Fig. 5 shows the brightness of the EL lamp made at the same conditions of printing and tested at the same parameters by using the additional dielectric layer with different dielectric constants. The additional dielectric layer increased the reliability of EL devices up to 25 %. However, the choice of polymer with a high dielectric constant is restricted.

Another way to increase the reliability of EL lamps lies in reaching a good electrical stability of the dielectric layer located between the phosphor layer and rear electrode. The dielectric layer is performed as a composite with a proper concentration of polymer binder and powder with high permittivity ($BaTiO_3$). The effective dielectric constant of this heterosystem as an EL lamp was described by the equation [5].

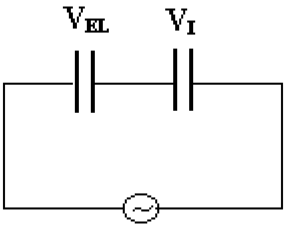

Fig. 4. Equivalent electric circuit of the EL cell.

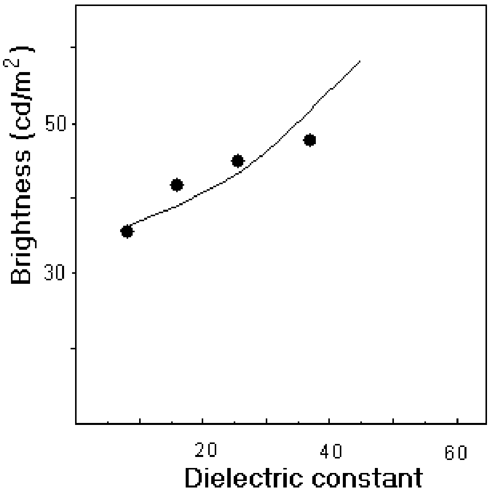

Fig. 5. Dependence of the brightness of EL lamp on the dielectric constant of additional dielectric layer.

$$\varepsilon_{\text{eff}} = \varepsilon_1\left[1 + 3f_2\,\frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2(1 - f_2) + \varepsilon_1(2 + f_2)}\right], \qquad (6)$$

where ε_1 is the dielectric constant of binder, ε_2 – dielectric constant of filler ($BaTiO_3$), f_2 – portion of filler in unit value (as concentration). The function (6) has no extreme, thus the optimum value f_2 should be experimentally selected.

The choice of a suitable concentration for the components allows to obtain an optimal dielectric constant of the insulating layer. To reach it, different concentrations of a high dielectric constant filler (powder $BaTiO_3$) at equal thicknesses of the phosphor layer were tested. The optimum concentration of the filling compound in view of the printing requirements was 70 % $BaTiO_3$ (Fig. 6). The reliability of samples was increased by 30 % using the second dielectric layer with 70 % $BaTiO_3$ (thickness 20 µm).

There is another possibility to increase reliability by using a high concentration of phosphor powder inside the phosphor layer. It corresponds to the decreasing distance between ZnS grains. If the phosphor powder and thin layer of binder between phosphor are considered as a series electric circuit of capacitors, than the capacitance of thin binder layer will increase despite the increasing concentration of phosphor powder. So, it means that the voltage drop in phosphor powder is increased. The brightness of an EL cell will be

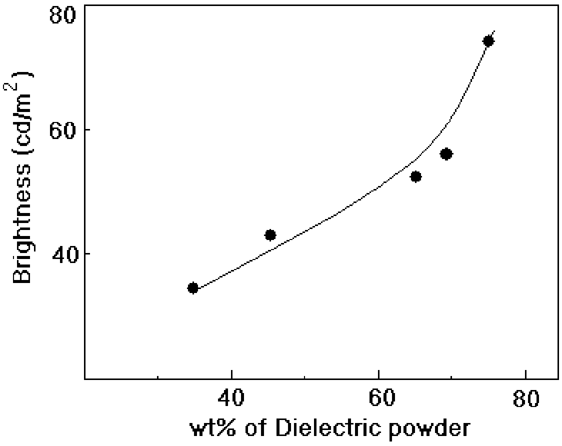

Fig. 6. Dependence of the brightness of EL lamp on the concentration of $BaTiO_3$ powder.

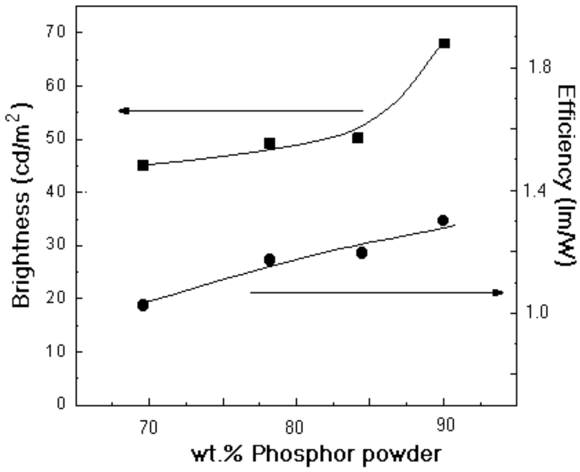

Fig. 7. Dependence of the brightness and efficiency on the concentration of phosphor powder.

increased, but using a thick dielectric layer it is possible to make initial brightness. At the same time, the increase of brightness did not change strongly the current of the EL cell, from where it follows that the efficiency is grown (Fig. 7).

The ideal variant, at which the brightness of the EL cell has its maximum, will take place when the phosphor layer completely consists of phosphor particles. However, in this case it is impossible to print the phosphor layer by low concentration of binder. Therefore, the optimum concentration of phosphor powder was determined as 70 %. But the brightness of the EL cell can be increased up to the best value by increasing the thickness of phosphor layer. With this purpose, we studied how the brightness of the EL cell depends on the thickness of phosphor layer when all the other construction parameters of this cell were fixed. We found that the optimum of the phosphor layer thickness is 38-45 µm with the mean diameter of phosphor particles 25 µm (Fig. 8). It is well known that for hexagonal packaging particles with diameter D, the

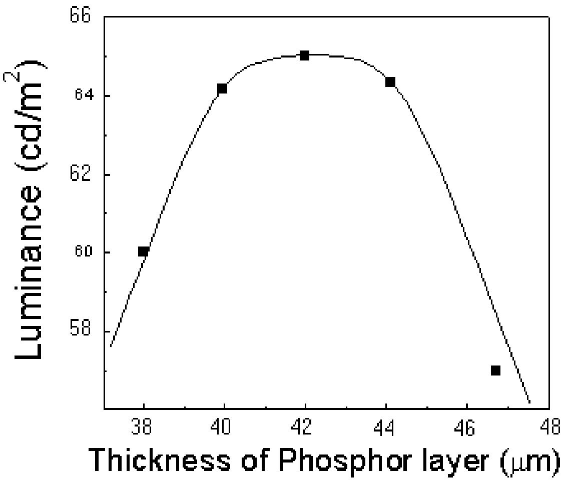

Fig. 8. Dependence of the brightness on the thickness of phosphor layer.

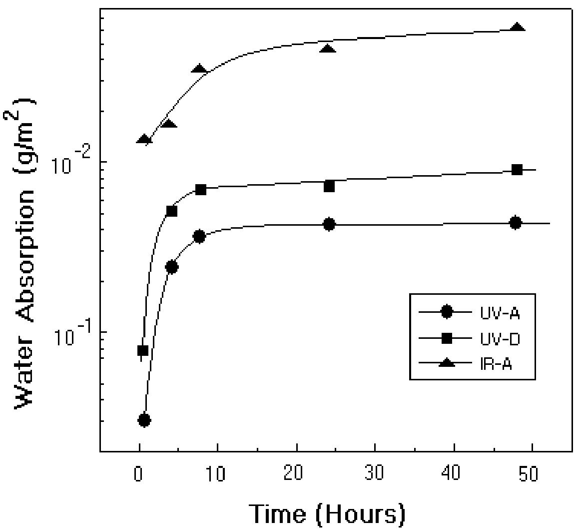

Fig. 9. The time dependence of water adsorption.

thickness of layer is $H = (1 + \sqrt{3}/2)D$. The simple calculations showed that the obtained thickness of phosphor layer satisfies the above condition. Consequently, the optimum packaging particles of phosphor is close to the hexagonal one.

Packaging the printed EL lamp layers is important for reliability. It was shown that the reliability of EL lamps strongly depends on waterproof properties of packaging material (Fig. 9). Testing various polymer materials, we found that, after a soon start, water adsorption was stabilized in 10 hours. For the first ten hours, water adsorption of tested UV polymer was 10 times increased. But the rate of water adsorption for IR polymer was 3 times lower than that for UV polymer. The reliability of EL cells packaged with IR ink was shown to be 10 % higher than for UV ink.

4. Conclusion

The obtained results are indicative of the following optimum ways and parameters to get the high brightness and reliability. One should use:

- balanced resistance between an electric circuit and an EL lamp;
- additional dielectric layer between the phosphor layer and transparent electrode;
- binder with a high dielectric constant;
- concentration of phosphor powder 70 % and binder 30 %;
- thickness of phosphor layer equal to $H = (1 + \sqrt{3}/2)D$, where D is the mean diameter of phosphor particles;
- package material with high waterproof properties (IR polymer).

It is necessary to mark in the conclusion that the given article does not allow to spot a nature of origin of failures, and most likely views methods of elimination them. These results will be useful for manufacturing and qualitative controlling the EL lamps.

Acknowledgments

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