SCIENTIFIC PAPERS
OF THE UNIVERSITY OF PARDUBICE
Series A
Faculty of Chemical Technology
19 (2013)

BEHAVIOUR OF PRINTED ELECTROLUMINISCENT PANEL IN ACCORDANCE TO ORDER OF LAYERS

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Received September 30, 2013

In printing industry it is an important step to create more interactive designs to attract the customers. One of them is the utilization of electroluminescence. The properties of an electroluminescent device depend on the panel's structure, used materials, driving voltage and frequency. In this paper, an all printed electroluminescent panel is presented with different layer orders or number of layers. The panels' structures differ in the position and thickness of the dielectric layer. These different structures are causing changes in threshold voltage, highest obtainable luminance and durability of the panel. The behaviour of the panels and crystals of the active layer was inspected via spectrophotometry and optical microscopy in the range of voltages from 10 to 120 V at frequencies of 50 and 500 Hz driven by an AC power source. The results show that the panel structure with a single dielectric layer from only one side of the active layer, independently on the position (in front of or behind the active layer) has the highest luminance. It was revealed that the best durability is achieved with a double dielectric layer from both sides of the active layer. Occurrence of sparking is not acceptable, and

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thus as the most compliant panel structure is the panel with one dielectric layer from both sides of the active layer with luminance of more than 20 cd m^{-2}*. The heat arising from the sheet resistance of indium tin oxide showed a slight red shift in the emission spectra. A test on a ceramic heating table revealed that the shift is caused by heat.*

Introduction

Relevance of utilization of electroluminescence in printed media is growing in the field of attracting consumers to printed products or in the field of producing displays for its lower price of production via printing processes.

The first task before printing an electroluminescent (EL) device is to choose the right electroluminescent material and to find the most convenient device structure. Behaviour of the created device is also important and needs to be studied before creating a functional device.

The ink formulation for preparation of electroluminescent layers is often based on electroluminescent doped phosphors such as ZnS. Electroluminescent behaviour in such crystals was first observed by Destriau in 1936 [1]. EL characteristics of ZnS differ in dependence on the used activators, co-activators and dopants. From that time there has been many advances in the field [2]; this study would contribute to the use of Cu doped ZnS in the field of printed applications. To do that, the aim of the paper is to describe and understand the behaviour of printed EL panels which are printed in different structures varying in layer order and thickness.

Materials

The printed panels use PET (polyethylene terephthalate) with a predeposited layer of indium tin oxide (ITO) as the substrate. ITO was chosen due to its high transmission that can reach even 90 % in the visible range and, therefore, it is compliant for use as the front electrode in an EL panel [3]. On the other hand, the disadvantage of ITO layer can be its sheet resistance [4]. Sheet resistance of the ITO on PET substrate is approximately 60 Ω sq^{-1} (determined with multimeter). The sheet resistance of this layer can be one of the reasons responsible for temperature changes of the panel as its temperature is rising with the increase of applied voltage. This temperature behaviour can affect the colour of emission and durability of the panel.

Another critical parameter for the proper function of an EL panel is the dielectric layer. The dielectric should possess a high dielectric constant to withstand dielectric breakdowns and the dielectric layers should be free of pin-holes. In this study a conventional screen-printing varnish Polyplast PY 383 from Sericol was used as the dielectric. Before use, the dielectric compound was mixed with Sericol ZV552 (drying inhibitor) in the ratio of 80 : 20 wt %.

The active layer was made of a commercially available compound of electroluminescent crystals from Metalor - Briflex Active Paste White/Blue. The electroluminescent ink formulation was modified with addition of butyl carbitol (diethylen glycol butyl ether) for ink drying inhibition in the ratio of 10 : 90 wt %. In one case for a panel without a single layer of dielectric, the formulation was modified with PY 383 varnish in the ratio of 20 : 80 wt %.

Silver ink 5029 DuPont was used as the rear electrode and for creating contact areas on ITO layer without any modification.

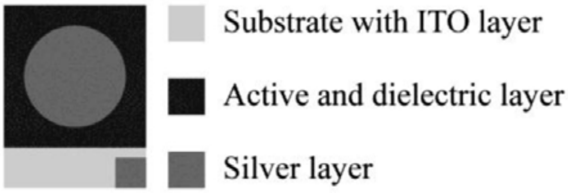
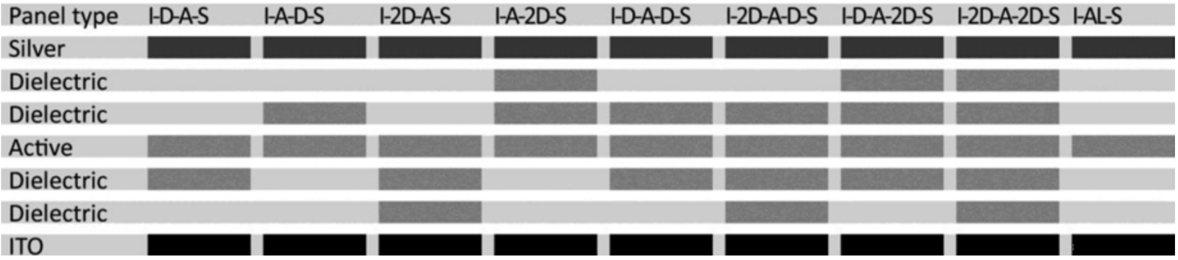


Fig. 1 Design of printed panels. First is the substrate with a predeposited ITO layer. The last is a silver conductive layer. Active and dielectric layers are sandwiched between these two layers in the structures shown in Fig. 2

Experimental

Printing of the electroluminescent panels was accomplished with screen printing with mesh count of 100 thr cm^{-1} (mesh tension 34 N). For printing, a semi-automatic screen printing machine (S-200 HF, Ever Bright Printing Machine FTY. Ltd.) was used. Design of the panel is depicted in Figure1, the panel types and their structure variations are described in Table I. Structure variations are illustrated in Fig. 2.



Panel type	I-D-A-S	I-A-D-S	I-2D-A-S	I-A-2D-S	I-D-A-D-S	I-2D-A-D-S	I-D-A-2D-S	I-2D-A-2D-S	I-AL-S
Silver	■	■	■	■	■	■	■	■	■
Dielectric				▨			▨	▨	
Dielectric		▨		▨	▨	▨	▨	▨	
Active	▨	▨	▨	▨	▨	▨	▨	▨	▨
Dielectric	▨		▨		▨	▨	▨	▨	
Dielectric			▨			▨		▨	
ITO	■	■	■	■	■	■	■	■	■

Fig. 2 Layer structures of printed electroluminescent panels. Darker colours represent the front and rear electrode, and the 50 % grey colour represents the sandwiched layers. The meaning of the abbreviations is explained in Tabe I

The printing substrate was cut to 2 x 3 cm pieces. All samples were washed in methanol for 2 minutes in ultrasonic bath right before the printing process.

As it was stated above, the electroluminescent ink formulation was modified with addition of butyl carbitol to slow down the drying. This inhibitor was not used for the structure I-AL-S where the EL compound was modified with PY 383 varnish. This type of active paste was used without any individual dielectric layer, because the used varnish is acting as the dielectric in this composition, separating the crystals from each other. In all studied structures, the active layer was printed twice in wet-to-wet printing system to acquire denser layers with a higher homogeneity of crystals distribution. Silver layer is also printed in the same way on every sample — printed once as the rear electrode.

Table I Printed panel types and layer structures

Panel type	Printed layers	Layer structure
I-D-A-S	3	ITO substrate - dielectric layer - active layer - silver layer
I-A-D-S	3	ITO substrate - active layer - dielectric layer - silver layer
I-2D-A-S	4	ITO substrate - 2× dielectric layer - active layer - silver layer
I-A-2D-S	4	ITO substrate - active layer - 2× dielectric layer - silver layer
I-D-A-D-S	4	ITO substrate - dielectric layer - active layer - dielectric layer - silver layer
I-2D-A-D-S	5	ITO substrate - 2× dielectric layer - active layer - dielectric layer - silver layer
I-D-A-2D-S	5	ITO substrate - dielectric layer - active layer - 2× dielectric layer - silver layer
I-2D-A-2D-S	6	ITO substrate - 2× dielectric layer - active layer - 2× dielectric layer - silver layer
I-AL-S	2	ITO substrate - active compound dispersed in dielectric layer - silver layer

Structures of the created panels vary by adding, removing, or doubling the dielectric layer. Drying of the dielectric layer was slowed down by addition of Sericol ZV552 acting as a drying retarder in the above stated ratio.

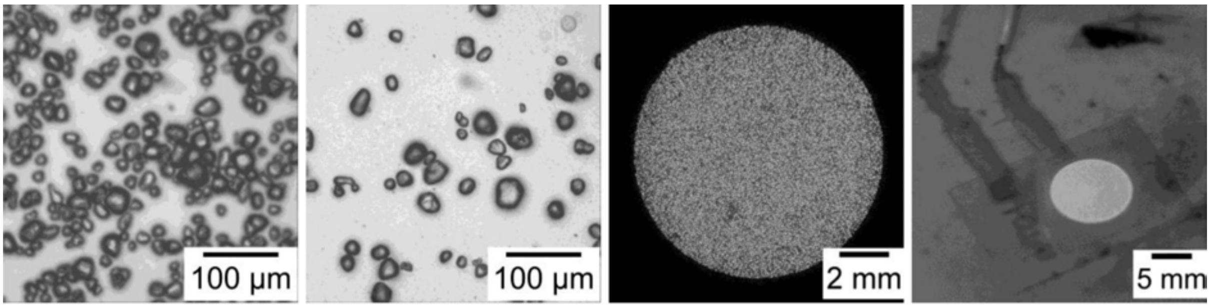
Panels were dried after each deposited layer. Drying of the dielectric layer was performed at room temperature in air. Time of drying of dielectric layer was set to 20 minutes at room temperature, other layers were dried for 10 minutes at 120 °C.

Finally, the samples were cut to their final size to fit in the contacting table. Mounted onto the table, the samples were driven by an AC power source and measured with spectrophotometer i1 Pro.

Except for the spectrophotometric measurements, the panels were also investigated with optical microscopy and thermocamera. The thermocamera was used to monitor the panels' temperature at various voltages. It was also used as a controlling device during the measurements of the influence of temperature on emission colour. The emissivity of the camera was measured using a plate of frosted glass placed on the heating table and was set to $\varepsilon = 0.96$.

Results and Discussion

The investigation of the panels with optical microscopy revealed some basic properties of the active layer and gave an opportunity to describe its behaviour. With optical microscopy, it could be observed that the crystal dimensions varied from 5 to 45 μm (Fig. 3a) which could influence the resultant luminance [5]. Typical coverage density of crystals for all panels except I-Al-S was approx. 1690 crystals mm^{-2}. This applies for all the panels except for panel I-AL-S. The density of crystals was much lower in this case, approx. 360 crystals mm^{-2} (Fig. 3b). These values were acquired from micrographs studied in ImageJ software (Wayne Rasband, version 1.46r) where the crystal count was calculated.



a) b) d) c)

Fig. 3 Active layer of panels a) applies to all panels but I-AL-S and b) applies to panel type I-AL-S. In c) an AC driven panel on ceramic heating table can be seen during testing of its behaviour at different temperatures, and d) is a macrophotography of the glowing crystals of the panel

The differences in particle dimensions can be responsible for affecting the endurance of the panel. As the particles differ in size, they create an uneven surface. The dielectric layer which is deposited on the active layer can then also be uneven in thickness. The electrical breakdowns could be caused by the presence of the larger crystals among the smaller ones. The inhomogeneity of the dielectric layer is due to the ink flowing off of the sticking crystals from the active layer. This could result in thinning of local areas of the dielectric layer, and the presence of such areas can consequently cause breakdowns when panels are driven by AC power.

Table II Obtained threshold (1 cd m^{-2}) voltages of printed panels at frequencies of 50 and 500 Hz. (Measurement error below 1 V)

Panel type	Threshold voltage, V	
	@ 50 Hz	@ 500 Hz
I-D-A-S	59	31
I-A-D-S	57	29
I-2D-A-S	64	33
I-A-2D-S	74	36
I-D-A-D-S	71	35
I-2D-A-D-S	97	45
I-D-A-2D-S	93	45
I-2D-A-2D-S	113	57
I-AL-S	> 120	45

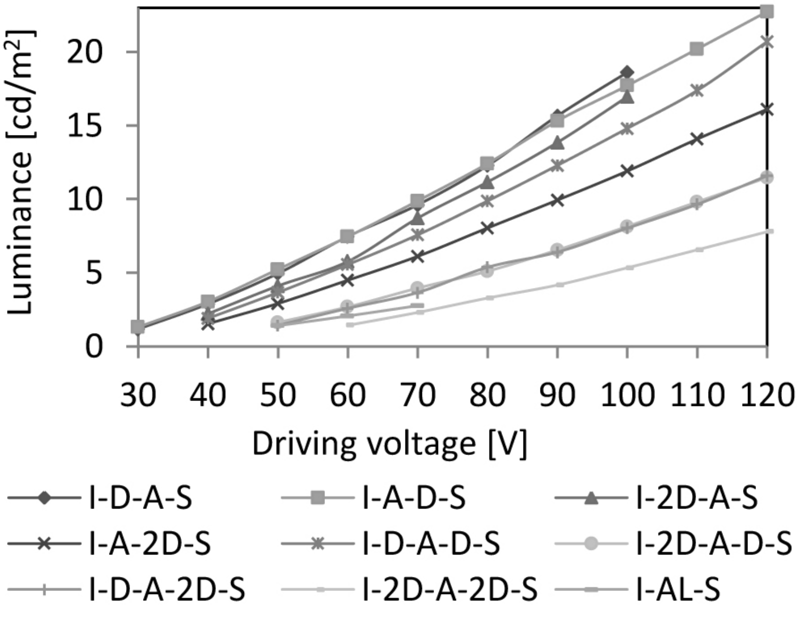


Fig 4 Obtained luminance according to the driving voltage at 500 Hz

On the other hand, the rough surface of the active layer can be advantageous in the case when the next layer is the silver conducting layer. The advantage arises from the fact that the luminance is increasing with the reducing distance of the electrode from the crystals. In this case the crystals are surrounded by the electrode and the silver electrode has even much lower sheet resistivity than ITO, approximately 0.015 $\Omega\,\mathrm{sq}^{-1}\mathrm{mil}^{-1}$ [6]. The effect of direct contact of the active layer with rear electrode can be seen in Fig. 4 where panel types I-D-A-S and I-2D-A-S are brighter than other panel types (except for I-A-D-S). Unfortunately, this higher luminance comes with lower endurance of the panel. Luminance in I-A-D-S and I-D-A-S structures is similar despite of the non-direct contact of the active layer with rear electrode. Here, the active layer is directly connected with the front electrode. Similarly, I-A-2D-S panel type has a direct connection of front electrode with active layer but the luminance is much lower than for the panel type with a direct connection of active layer with the rear electrode.

During observation of the behaviour of crystals through microscope, differences in colour emission of individual crystals could be seen. Some crystals emit only in green range of spectrum and the other crystals in blue. On decreasing the frequency from 500 to 50 Hz, all crystals change their emission colour to green.

At voltages over 60 V (at 500 Hz frequency), one could distinguish that the highest luminance arises from the edges of crystals. It can also be seen that the smooth sides of crystals do not emit any radiation but if such a crystal side contains a defect in the crystal structure, light is obtained from that place and can be even more intense than the one obtained from the crystal edges. This high luminance in the defect area can be due to a high specific surface area which forms the defect and thus appears brighter than the rest of the crystal.

This finding implies that the highest luminance can be obtained from a crystal with a rough surface and high specific surface area. That way the crystal would have many edges and rips capable of light emitting.

With optical microscopy it is possible to observe that before a breakdown of a crystal in I-2D-A-2D-S panel type, the crystal starts to flash. The flashing persists to the point of its resistance to breakdown at a particular applied voltage. Before the breakdown itself, the crystal starts to emit a violet-to-white light. Sparking of the crystals can be observed more often in areas of crystal close to the electrode. After a breakdown the whole crystal ceases to glow thus creating a local area without electroluminescence in the panel. Sparking affects mostly the blue emitting crystals. It was observed that sparking can occur since driving a panel from threshold voltage. But mostly, this sparking started at higher voltage values, and the rate of sparking increased with increasing driving voltage. In a case when the panel is turned off at room temperature and turned on using higher driving voltages, e.g., 100 V, the panel undergoes a shock and the whole panel can get damaged and unusable. On the other hand, if a panel is once powered from its threshold to its highest limits of driving voltage, the sparking will not appear at all or at least the rate of sparks will not be as high as during the first testing. Observation of sparks could be achieved only with double dielectric layers from both sides of the active layer. With less dielectric layers the observation was more complicated because the breakdown process in panels is much faster, and it cannot be predicted which crystal will break down.

To test the endurance of panels, voltage range from 0 to 120 V was used. According to the layer structure of panels it is obvious that I-AL-S panel type will not withstand higher voltages and will have the worst results of all panels tested. Breakdown of crystals was controlled only by visual observation. The results of the highest applicable driving voltages were assigned according to the tester's consideration. During testing, increasing the applied voltage, sparks were observed and sometimes even accompanied by crisping sounds. If those sparks and crisps were frequent, and the panel was considered not to withstand any further increase of voltage, then the testing was terminated.

The results of endurance of the panels revealed that the structures without a dielectric layer between the active and rear (silver) electrode failed as the first in testing. Two panels, I-D-A-S and I-2D-A-S, did not withstand driving voltages higher than 110 V. The sparking and crispy sounds were occurring from the very low driving voltages (around 30 V). At more than 100 V the sparking was unbearable and the measurement had to be terminated. The worst results were obtained for I-AL-S panel type where the driving voltage could not be increased above 70 V due to high sparking rate. Other panel types withstood more than 120 V with low frequency or no sparking. I-2D-A-2D-S panel type started to spark

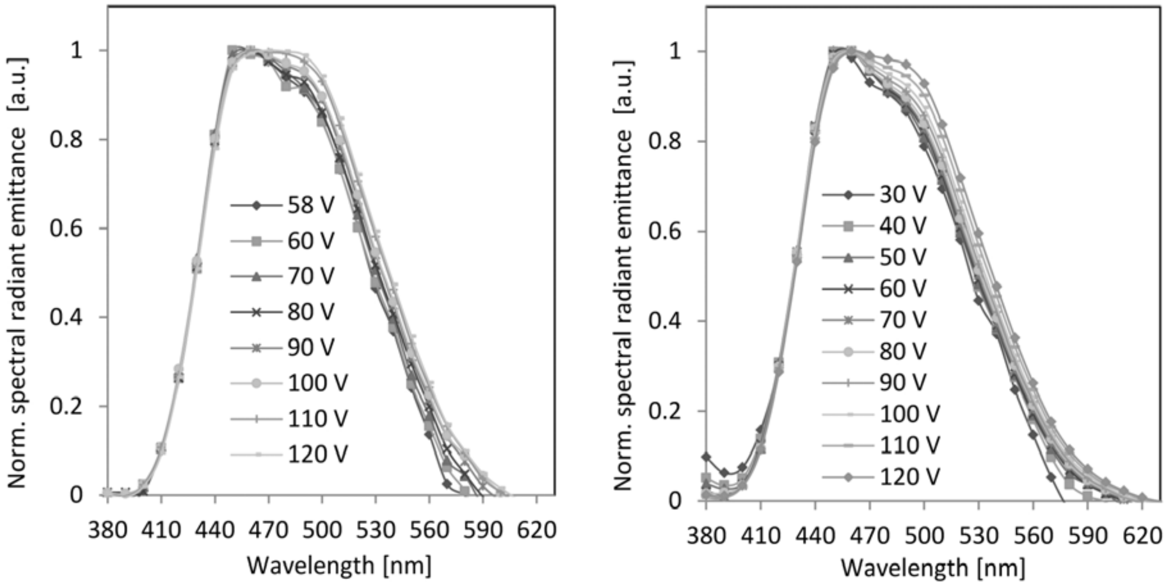


Fig. 5 Normalized spectral radiant emittance of panel a) I-2D-A-2D-S and b) I-A-D-S from threshold voltage to 120 V at 500 Hz. The emission colour is broadening to higher wavelengths and the intensity at 460 nm is quenched with the increase of driving voltage

above 150 V and withstood even 180 V. The sparking at driving voltage close to 180 V was too frequent and the panel's temperature raised over 100 °C. The temperature influenced the panel's emission colour which is evident from Fig. 5. The cause of the spectral shift could arise from the heat released from ITO layer due to its sheet resistance.

According to the obtained results of colour change, other experiment was performed. Here, the spectral distribution of emission intensity was tested according to the applied temperature. A ceramic heating table was used for testing on which two silver conducting paths were directly drawn to retain the smallest gap between the heating table and panel. During the experiment, a constant driving voltage of 50 V was set, and the temperature was slowly increased by a ceramic heating table (Fig. 3c). The temperature was controlled with a thermocamera. The luminance of all stated variable panel structures from Table I was investigated in the same range of driving voltages. The obtained results (Table III) show that I-A-D-S panel type is the brightest of all at the maximum voltage (Fig. 4). However, this panel type showed some sparking during testing. Due to this finding I-D-A-D-S panel type, with the second highest luminance, is a better candidate for utilization. This panel type is protected from both sides of the active layer with a single dielectric layer resulting in higher durability. There was no sparking observed in the whole measured range. I-D-A-S, I-2D-A-S and I-AL-S panel structures were discarded from this comparison due to their low endurance. None of the obtained luminance is sufficient according to the standard luminance defined by ANSI [7].

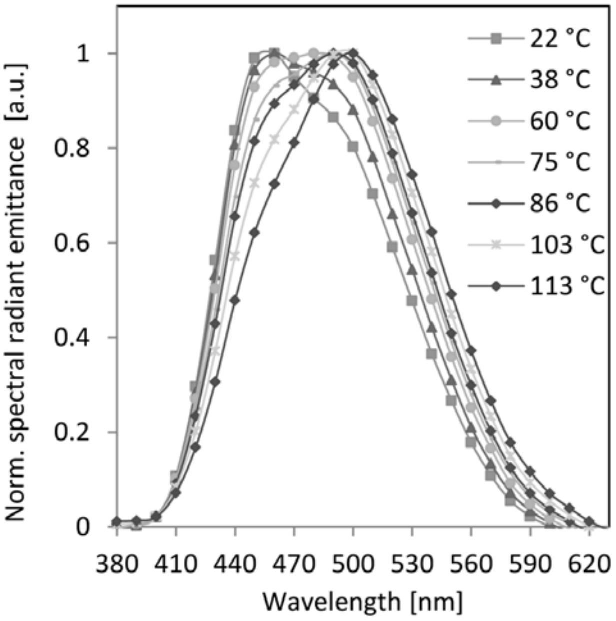


Fig. 6 Influence of temperature on spectral distribution of spectral radiant emittance of I-A-D-S panel type at 50 V and 500 Hz. Emittance at 460 nm is gradually quenched according to the applied temperature

As it can be seen in Fig. 6, the emission spectrum is not only broadened but also a decrease in intensity of radiant emittance around 460 nm is occurring. On the other hand, radiant emittance increase is taking place at around 500 nm with increasing temperature. If only the two temperature extremes are compared, a shift of the spectral distribution can be seen. When considering the whole spectrum, there is not one but two EL bands that are temperature dependent. If the temperature of the panel is raised, then quenching of the luminescence around 460 nm is taking place. EL band peak has shifted from 460 to 500 nm resulting in colour change from blue to green. Thus by heating the panel the results are similar to those when the temperature is raised due to the applied voltage. The heat (temperature increase) has also affected the intensity of luminance, changing it from 5.5 to 12.4 cd m^{-2} for temperature range from 22 to 113 °C, respectively.

The results shown in Fig. 5 are also connected with the quenching of luminance at 460 nm. Thus the temperature raise of the panel is causing the colour change. In this case the luminance is not quenched in such an amount as in Fig. 6 because the temperature, which could originate from sheet resistance of ITO, did not increase to comparable values.

Table III Luminance of panel types according to the applied voltage at 500 Hz. (Measurement error below 0.03 cd m^{-2})

Driving voltage V	Panel type and its luminance, cd m^{-2}								
	I-D-A-S	I-A-D-S	I-2D-A-S	I-A-2D-S	I-D-A-D-S	I-2D-A-D-S	I-D-A-2D-S	I-2D-A-2D-S	I-AL-S
30	1.17	1.31	-	-	-	-	-	-	-
40	2.88	3.04	2.21	1.54	1.90	-	-	-	-
50	4.95	5.23	4.10	2.91	3.66	1.63	1.42	-	1.40
60	7.46	7.45	5.77	4.49	5.55	2.68	2.58	1.43	2.06
70	9.61	9.88	8.71	6.11	7.57	3.95	3.65	2.30	2.76
80	12.28	12.42	11.16	8.04	9.88	5.10	5.37	3.28	-
90	15.66	15.31	13.85	9.93	12.29	5.23	6.40	4.16	-
100	18.61	17.71	16.96	11.91	14.81	8.15	8.03	5.33	-
110	-	20.19	-	14.08	17.38	9.81	9.64	6.55	-
120	-	22.73	-	16.10	20.70	11.46	11.58	7.78	-

The threshold voltage at the luminance of 1 cd m^{-2} [8] shows the dependence of panel's layer structure on driving voltage required for panel driving. The lowest voltage should be and is in the case of panel type with one dielectric layer used in the panel structure. On the other hand, the panel structure using dielectric layer combined with active layer shows its threshold higher than that with one separate dielectric layer. This is probably caused by lower density of active crystals in the layer. The obtained threshold values are shown in Table II for all printed panel types for frequencies of 50 and 500 Hz. For I-AL-S panel type, there is no threshold at 50 Hz frequency, because the demanded threshold voltage would be higher than 120 V and the panel was not able to withstand driving voltages higher than 70 V.

The distribution of the spectral radiant emittance (SRE) is changing with the applied voltage as it can be seen in Fig. 5. Normalized distributions of SRE are shown here for I-A-D-S structure. At 500 nm, the intensity is increasing with the increasing applied voltage resulting in a broad peak of SRE which changes the final emission colour. If similar intensities of luminance of all panel types are compared, the result is very alike, because for different panel types a different driving voltage had to be chosen (Fig. 7). The panels' temperature is changing with the driving voltage and, therefore, the spectral distribution of their SRE can be presented in accordance to the particular driving voltage (or temperature).

If the AC voltage frequency is lowered to 50 Hz, the shifts in spectra are not that apparent (Fig. 8). The EL band remains similar under any driving voltage from the specified range.

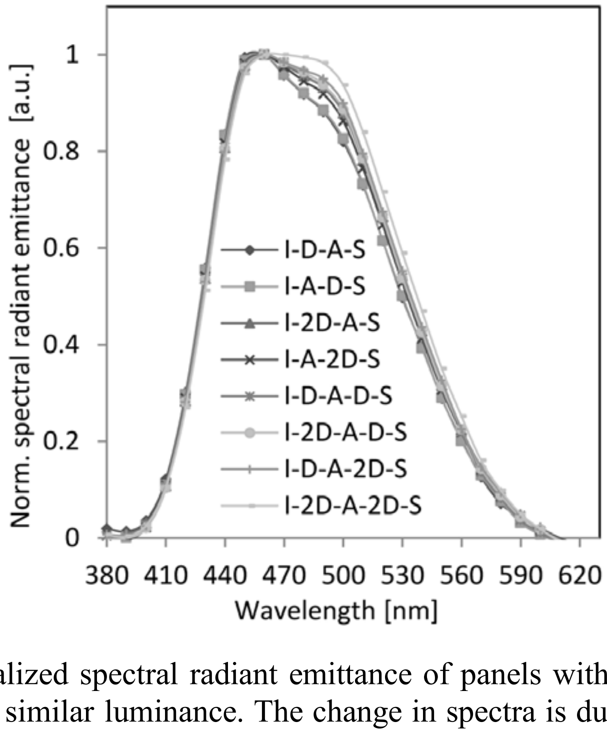


Fig. 7 Normalized spectral radiant emittance of panels with different structures with nearly similar luminance. The change in spectra is due to different AC driving voltage used for obtaining a similar emittance value. Used voltages were as follows: 60 V for I-D-A-S and I-A-D-S, 70 V for I-2D-A-S and I-D-A-D-S, 80 V for I-A-2D-S, 100 V for I-2D-A-D-S and I-D-A-2D-S, and 120 V for I-2D-A-2D-S at 500 Hz

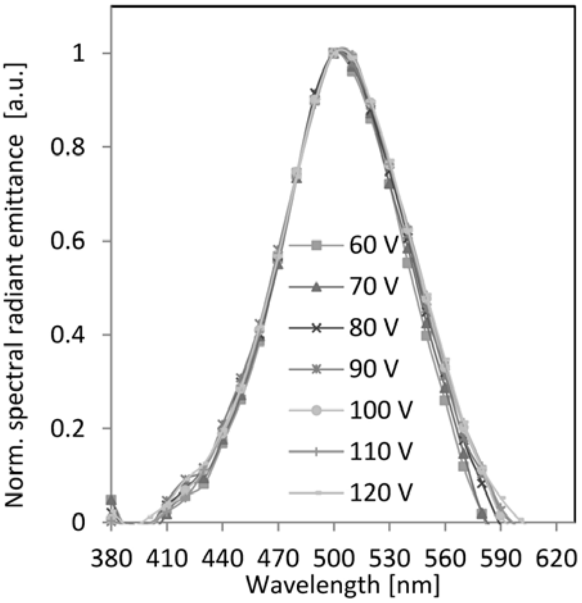


Fig. 8 Spectra of I-A-D-S panel type at different driving voltages at 50 Hz

Conclusion

In this paper, the behaviour of electroluminescent panels with various layer structures was investigated. Distribution of spectral radiant emittance and its intensity of these panels are dependent on many factors. First of all it depends on the used electroluminescent compound itself. Distribution of EL and its intensity is controlled by the applied voltage, frequency and even the temperature of the panel. The third factor belongs to the worst controllable ones because it is mostly unpredictable in common environments.

According to the endurance results, panel with two double layers of dielectric (I-2D-A-2D-S) is the best choice for withstanding higher voltages. On the other hand, the use of such thick dielectric layers increases the demand on driving voltage and lowers the obtainable luminance. In the tested voltage range it was found that panel type with two single dielectric layers is a sufficient device structure.

From luminance values it is clear that the best results were obtained for I-A-D-S structure with more than 22 cd m^{-2} at 120 V and 500 Hz. Unfortunately, this panel showed its limitations in durability against breakdowns as lower than in structures with dielectric layers from both sides of the active layer. Closest to the performance of I-A-D-S panel type is I-D-A-D-S with luminance more than 20 cd m^{-2} (at 120 V and 500 Hz). This panel type showed no sparking in the whole voltage range.

By increasing the driving voltage it was shown that also the temperature of the panel is increasing. This increase could be caused by sheet resistance of ITO. The temperature caused quenching of luminance around 460 nm resulting in colour change from blue to green.

The best suitable panel structure according to overall functional properties is the panel type with a single dielectric layer at both sides of the active layer (I-D-A-D-S). Unfortunately, the obtained luminance of every panel structure is very low for practical utilization.

There are several adjustments which could enhance the luminance of the panel. The active layer should be composed of crystals with narrow distribution of size of EL particles. Thus preventing other layers to be deposited on an inhomogeneous surface, i.e., from which the dielectric paste could flow of and distribute itself unevenly. A higher coverage density could be improved by choosing a screen mesh with lower density. The dielectric layer could be modified or changed for other with better characteristics.

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