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MODEL OF AGEING PROCESS OF ELECTROLUMINESCENT STRUCTURES*

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In the presented paper the authors propose a new mathematical model of aging process proceeded in thick film electroluminescent structures. The impact of humidity, temperature and supply voltage parameters (amplitude, frequency) on luminance changes during long time exploitation tests was taken into consideration. It was observed that the electroluminophore in Destriau electroluminescent cells exhibit very fast decrease in luminance in the initial phase of exploitation. The results of investigations indicate that the increase of amplitude and frequency of supply voltage accelerate the aging process. The rate of the aging process is also significantly influenced by the ambient temperature and elevated humidity combined with a supply voltage. The proposed model enables to estimate the life-time of electroluminescent structures on a base of exploitation tests in their initial stage.

Keywords: electroluminescence, ageing process, mathematical model

1. INTRODUCTION

Electroluminescence is a phenomenon of light emission, which takes place in a luminescent material exposed to an electric field [1].

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The electroluminescent structures are more and more widely applied, as the light emitted by them is characterised by low power consumption, uniform emission from the whole active surface, the possibility of continuously variable control of light .

These structures are however subject to the ageing process, the rate of which depends upon many factors. Under the influence of the electric field applied and climatic operating conditions, i.e. temperature and humidity, irreversible changes take place in the electroluminophore structure that result in a systematic decrease in luminance of the light emitted. In order to propose a model of the ageing process of electroluminescent structures and determine the dynamics of luminance change processes, the authors have screen-printed the thick-film electroluminescent structures on flexible substrates and carried out a series of operation tests where the major parameter assessed was the luminance value.

2. SAMPLE PREPARATION

In Fig. 1, a schematic diagram of the sandwich electroluminescent structures composed of screen-printed layers is presented.

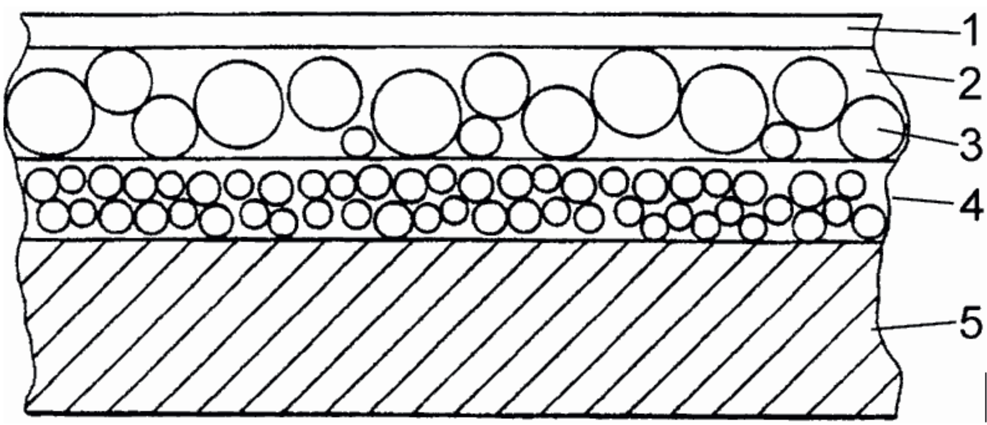

Fig. 1. Arrangement of layers in an electroluminescent structure: 1 – polyester foil with an ITO film; 2 – transparent thermosetting resin; 3 – electroluminophore grains; 4 – dielectric layer; 5 – conductive layer (opaque).
Rys. 1. Układ warstw w strukturze elektroluminescencyjnej: 1– folia poliestrowa z warstwą ITO; 2 – przeźroczysta żywica termoutwardzalna; 3 – ziarna elektroluminoforu, 4 – warstwa dielektryczna; 5 – warstwa przewodząca.

The following polymer ink films have subsequently been deposited onto the polyester substrates of the dimensions of 30 mm x 20 mm, pre-coated with an ITO

(*Indium Tin Oxide*) layer of a sheet resistance of 400Ω/: an electroluminophore film whose major component are the grains composed of the Cu-activated and Cl--co-activated ZnS crystals suspended in the resin, a dielectric layer composed of the $BaTiO_3$ powders also suspended in the resin, and a conductive layer, a composition composed of silver powders and thermosetting resin. After deposition, each layer was cured at a temperature of approx. 130°C for 30 minutes. After the curing processes have been completed, the structure underwent visual inspection and luminance measurements were subsequently carried out at the a.c. voltage with an amplitude value of 100 V at a frequency of 400 Hz. The luminance measurements were performed with a Tektronix J18 luminance meter and the error of measurement did not exceed 5%.

3. TEST PROCEDURES

The following tests have been carried out on fabricated samples:

- a reliability test at the temperatures of +20°C, +40°C, +60°C, at a supply voltage U_{p-p} = 180 V, f = 400 Hz. The results of the test are shown in Fig. 2,

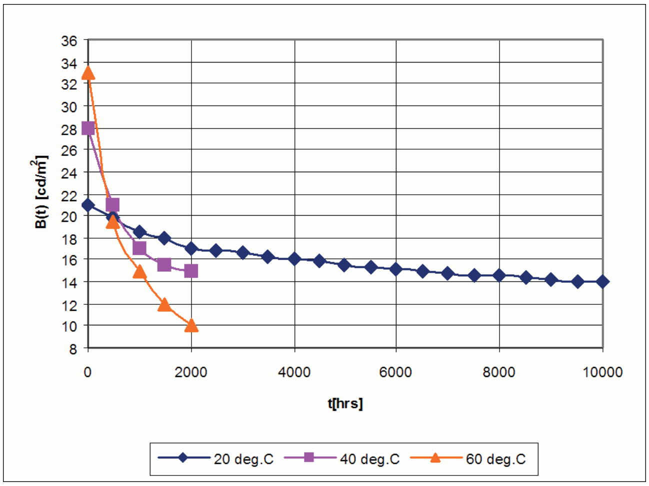

Fig. 2. Changes of luminance during reliability tests.
Rys. 2. Zmiany luminancji podczas badań niezawodności.

- a reliability test at the ambient temperature of +20°C, at variable supply voltages and a constant frequency 400 Hz. The results of the test are shown in Fig. 3,
- a reliability test at the ambient temperature of +20°C, at variable frequency and a constant supply voltage 150 V. The results of the test are shown in Fig. 4,
- a moisture resistance test under conditions of the prolonged exposure to moisture

at a temperature of +40°C and a relative humidity of 98%. The results of the test are shown in Fig. 5.

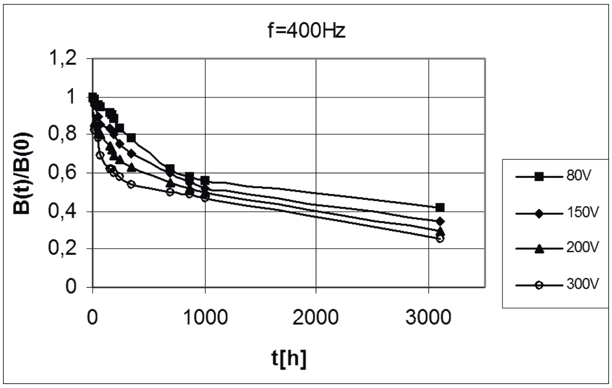

Fig. 3. Results of exploitation tests at constant frequency of supply voltage.
Rys. 3. Wyniki badań eksploatacyjnych przy stałej częstotliwości napięcia zasilającego.

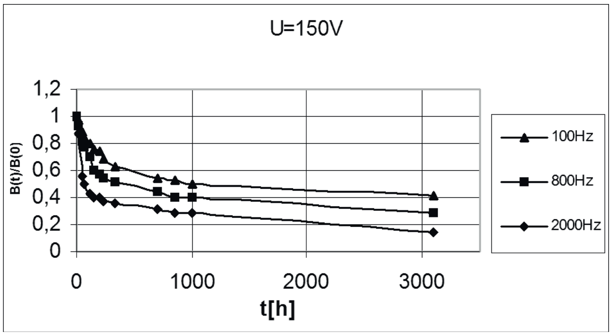

Fig. 4. Results of exploitation tests at constant supply voltage.
Rys. 4. Wyniki badań eksploatacyjnych przy stałym napięciu zasilania.

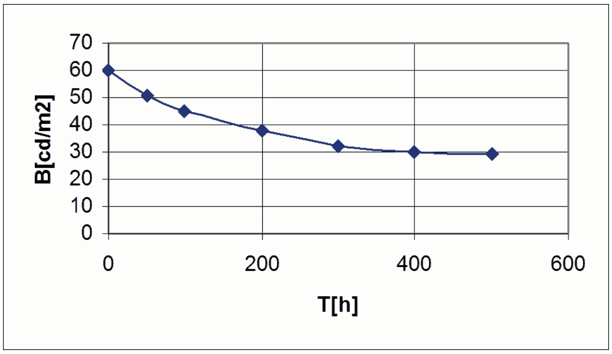

Fig. 5. Changes of luminance during humidity tests.
Rys. 5. Zmiany luminancji podczas prób odporności na wilgoć.

The results of the tests conducted indicate that one of the most important factors that cause the deterioration of the electroluminescent structures is humidity, which penetrates the luminophore grains and performs the role of an ion conducting medium, and causes the decomposition of ZnS [2] so that it loses its semiconductor properties. The process is particularly intensive in the presence of electric field.

4. A GENERALISED MATHEMATICAL MODEL FOR AGEING PROCESSES

Several independent phenomenons take place in the general ageing process, two of which play an important role: the decomposition of luminescence centres under the impact of moisture and the other much slower process — the decomposition of the ZnS grains due to the reaction with copper and consequently the precipitation of metallic zinc according to the reaction :

$$ZnS + Cu = CuS + Zn$$

In both the processes, the concentration of luminescence centres n decreases what can be written in the form of the following equation:

$$\frac{dn}{dt} = -\frac{n}{\tau} \tag{1}$$

where τ is a time constant of the process.

The solution to the equation (1) has a general form:

$$n(t) = n(0)\exp(-t/\tau) \tag{2}$$

As the luminance B of an electroluminescent film is directly proportional to concentration of active luminance centres [1], the following formula can be written:

$$B(t)/B_0 = a_1 \exp(-t/\tau_1) + a_2 \exp(-t/\tau_2) \tag{3}$$

where: $B(t)$ is the instantaneous value of luminance, B_0 is the luminance at the initial moment of time and a_1 is a numerical coefficient connected with the luminance centre deterioration process under the impact of moisture, τ_1 – is a time constant of this process, while a_2 is an analogous to a_1 coefficient connected with the decomposition of the ZnS grains, and τ_2 is a time constant of this process, in which:

$$a_1 + a_2 = 1 \tag{4}$$

The values of the τ_1 and τ_2 time constants decrease with increasing applied voltage U and increasing temperature (ΔT – the increase in temperature against the room temperature), because the structure ageing process accelerates the increase in operating temperature. This can written in a simplified form:

$$\tau_1 = \frac{\tau_{01}}{1 + b_1 U + c_1 \Delta T} \quad (5)$$

and

$$\tau_2 = \frac{\tau_{02}}{1 + b_2 U + c_2 \Delta T} \quad (6$$

where τ_{01} and τ_{02} are the time constants for the first process and the second one, respectively, at the room temperature when no voltage is supplied, b_1 and c_1, and b_2 and c_2 are the constants for the first process and the second one, respectively.

The reliability tests have showed that the increase in frequency of the supply power (Fig. 4) causes a faster deterioration of the structure than the increase in amplitude of the supply voltage (Fig. 3). At an supply voltage having an effective value of 150 V, a change in frequency from 100 Hz to 2000 Hz causes over 10-fold increase in the rate of the ageing process, while the change in supply voltage from 80 V to 300 V results in only 2-fold acceleration of this process (at a frequency of 400 Hz). This provides evidence that it is not as much the length of a period of time the structures are exposed to the voltage as the number of change cycles (N) of the sinusoidal supply voltage (with a frequency f) applied to a structure, i.e. t = N/f [3], that have the decisive impact on the change in luminance. Thus, at a fixed number of cycles N, the τ_1 and τ_2 time constants are inversely proportional to frequency.

On the basis of the investigations carried out and the calculations performed, one may accept that the proposed model satisfactorily describes the ageing processes in the electroluminescent structures, and the parameters of the model determined based on the measurement results (for a chosen structure) have the following values:

a_1 = 0,3; b_1 = 0,016 V^{-1};
c_1 = 0,13 K^{-1}; τ_{01} = 590 h,

for the deterioration process of the luminance centres under the impact of moisture, and

a_2 = 0,7; b_2 = 0,007 V^{-1};
c_2 = 0,1 K^{-1}; τ_{02} = 7100 h,

for the ageing process connected with the decomposition of the ZnS grains and the precipitation of metallic zinc.

Discrepancies between the values calculated based on the proposed model and those measured are of the order of a few per cent, and they do not exceed 10% in the worst case.

5. CONCLUSIONS

It results from the investigations carried out and the calculations performed that the proposed mathematical model satisfactorily describes the ageing processes in the electroluminescent structures with the equations (1) to (6), and it is possible on the basis of the measurement results to determine the model parameters for a chosen structure.

Based on the investigations results (Fig. 2. to Fig. 5.) and calculated parameters of ageing model it is evident, that moisture and elevated working temperature in the highest degree influence the ageing of electroluminescent structures.

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MODEL PROCESU STARZENIA STRUKTUR ELEKTROUMINESCENCYJNYCH

Autorzy proponują w artykule nowy matematyczny model procesu starzenia zachodzącego w grubowarstwowych strukturach elektroluminescencyjnych. Uwzględniono wpływ wilgoci, temperatury i parametrów napięcia zasilającego (amplituda, częstotliwość) na zmiany luminancji podczas prób eksploatacyjnych. Zaobserwowano, że w początkowej fazie eksploatacji elektroluminofor w strukturach Destriau – warstwowych strukturach elektroluminescencyjnych, bardzo szybko zmniejsza luminancję. Wyniki badań wskazują, że wzrost wielkości amplitudy napięcia zasilającego i częstotliwości przyspiesza proces starzenia. Szybkość procesu starzenia struktur elektroluminescencyjnych w istotny sposób zależy od temperatury otoczenia i podwyższonej wilgotności w powiązaniu z napięciem zasilania. Zaproponowany model umożliwia oszacowanie czasu życia struktur elektroluminescencyjnych w oparciu o wyniki badań eksploatacyjnych, już w początkowej ich fazie.

Słowa kluczowe: elektroluminescencja, proces starzenia, model matematyczny