Reliability and Accelerated Test Methods for Plastic Materials in LED-Based Products

M. Yazdan Mehr\textsuperscript{1,2*}, W.D. van Driel\textsuperscript{2,3}, G.Q. Zhang\textsuperscript{2}

\textsuperscript{1} Materials innovation institute (M2i), Delft, The Netherlands
\textsuperscript{2} Delft University of Technology, EEMCS Faculty, Delft, The Netherlands
\textsuperscript{3} Philips Lighting, Eindhoven, The Netherlands

Abstract:
In this study the effects of thermal ageing on the optical properties of both lens and the remote-phosphor samples, made from Bisphenol-A polycarbonate (BPA-PC) are investigated. The BPA-PC lens and remote phosphor plates are currently widely used in light conversion carriers and optical lenses in LED-based products. Lens and the remote phosphor BPA-PC samples of 3 mm thickness were thermally aged at temperature range 100 to 140 °C. The phosphor plates, combined with a blue LED light source, produce white light with a correlated colour temperature (CCT) of 4000 K. The colour shifting due to thermal ageing was studied by Integrated Sphere. Results show that thermal ageing leads to a significant decrease in the luminous flux and chromatic properties of plates. It is also shown that by increasing the temperature, the kinetics of degradation reaction becomes faster, inferring that lumen depreciation takes place at shorter time. Lumen depreciation up to 30% reduction is extrapolated to temperatures lower than 100 °C. It is shown that the lifetime, defined as 30% lumen depreciation at 40 °C, is around 35 khrs for remote phosphor and around 100 khrs for BPA-PC lens. A significant change both in the correlated colour temperature (CCT) and in the chromaticity coordinates (CIE x,y) is also observed in thermally aged specimens. Deterioration of the chromatic properties of the phosphor plates is correlated to the decrease in the luminous flux. Results also confirm the colour shifting of white light towards yellow region. Based on the observed decay of CCT and colour shifting, one could conclude that the thermal degradation of the remote phosphor plates affects both the efficiency and the colour of the LED products. The proposed thermal-ageing qualification method can be used by industries to efficiently select the proper phosphor materials and verify the product design, without many trial-error based interactions.

Introduction:
The BPA-PC lens and remote phosphor plates are currently widely used in light conversion carriers and optical lenses in LED-based products [1- 4]. In 1996, a new lighting device was invented by Nichia Chemical Co. which is a blue InGaN LED chip coated with yttrium aluminum garnet yellow phosphor (Y3Al5O12:Ce, YAG:Ce). When the chip is biased under certain current, blue light is emitted by the InGaN chip through electron–hole recombination in the P–N junctions. Some of the blue light from the LED excites the YAG:Ce phosphor to emit yellow light, and then the rest of the blue light is mixed with the yellow light to generate white light [5].

To describe “colour”, chromaticity coordinates (x,y) and colour temperature (CT) are more often used, with the former being defined by the Commission Internationale de l’Eclairage (CIE) System and the latter being the temperature of a blackbody whose chromaticity most nearly resembles that of a light source [5]. Low colour temperature implies warm while high colour temperature appears to be a colder (more blue) light. Daylight has a rather low colour temperature when the sun is dawn, and a higher one during the day.

The main target of this paper is to investigate the effect of ageing temperature on the optical properties and the reliability of remote phosphor. For this reason a set of accelerated thermal stress tests were applied with temperature level between 100 and 140 °C. The reliability studies and life
time assessment at temperatures lower than 100 °C are done by extrapolation.

2. Experimental methods:

One type of 3 mm-thick remote phosphor plate with Correlated Colour Temperature (CCT) of 3000 K are used in this study. The specimens are kept in a furnace at 100, 120, and 140 °C up to 3000 h. Testing temperatures for accelerated lumen depreciation test is determined in such a way that the temperature does not exceed the glass transition temperature of the plastics. Glass transition temperature (Tg) of BPA-PC is 150 °C, so the maximum accelerated temperature is chosen 10 °C below the Tg. Optical properties of thermally-aged plates, i.e. Luminous flux depreciation, were studied at room temperature, using an integrated sphere.

3. Reliability model:

The reliability model for the life time assessment is based on an exponential luminous decay equation, where the time-to-failure can be calculated as [10]

$$\Phi(t) = \beta \exp(-\alpha t)$$  \hspace{1cm} (1),

where $\Phi(t)$ represents the lumen output, $\alpha$ is the rate of reaction or depreciation rate parameter, $t$ is time and $\beta$ is a pre-factor. According to alliance for solid state illumination system and technology (ASSIST), when lumen output, $\Phi$, is equal to 70%, $t$ is time-to-failure [6]. The rate of reaction, $\alpha$, is related to the activation energy of the reaction and to the ageing temperature as follows [7]

$$\alpha = A \exp\left(-\frac{E_a}{K T}\right)$$  \hspace{1cm} (2),

where $A$ is a pre-exponential factor, $E_a$ is the activation energy (ev) of the degradation reaction, $K$ is the gas constant, and $T$ is the absolute temperature (K).

4. Results:

It is noticeable that there is a significant decay both in the phosphor yellow emission and in the blue peak. As is shown in our previous work the yellowing of BPA-PC plates leads to the reduction in the light transmissivity of plates [8]. Reduction in yellow emission also illustrates the decay of phosphor conversion efficiency, Figure 1.

Figure 1: The evolution of spectral power distribution (SPD) of sample B at 140 °C

A more explanation of the effects of thermal-ageing on the performance of remote is given in Figure 2. This Figure shows the progress of the normalized flux intensity and as a result the degradation kinetics of the phosphor plates. Obviously, the degradation rate shows a significant dependence on the stress temperature level; the higher the ageing temperature, the higher the lumen depreciation and the degradation kinetics.

Figure 2: Normalized flux of remote phosphor plates at different thermal stress tests

The activation energy of the degradation reaction in LEDs depends on the materials and the working conditions. The activation energy can be calculated from Equation (2). In order to obtain the activation energy, the natural logarithm of the
reaction rates is plotted against the inverse of the absolute temperature (see Figure 3). The slope is multiplied by the negative of the gas constant to obtain the activation energy, \( E_a \), in the eV. Activation energy for remote phosphor is between 0.333 eV, which is in agreement with previous reported values [7].

Figure 3: Plot of ln (α) vs E/KT for remote phosphor

Thermal stress test also has some significant effects on the CCT. In Table 1 the variation of CCT during high temperature stress test is shown for remote phosphor plate. It is obvious that by increasing the thermal ageing time and the temperature, the CCT decreases. One can also notice that the higher the ageing temperature, the higher the degradation kinetics.

Table 1: Correlated colour temperature (CCT) during high thermal-stress tests (temperature is in degree C and time is in hour)

<table>
<thead>
<tr>
<th>T (°C)</th>
<th>t (hrs)</th>
<th>0</th>
<th>240</th>
<th>480</th>
<th>720</th>
<th>1440</th>
<th>2040</th>
<th>2880</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>3170</td>
<td>3167</td>
<td>3150</td>
<td>3128</td>
<td>3105</td>
<td>3090</td>
<td>3056</td>
<td></td>
</tr>
<tr>
<td>120</td>
<td>3170</td>
<td>3150</td>
<td>3120</td>
<td>3105</td>
<td>3065</td>
<td>3048</td>
<td>3018</td>
<td></td>
</tr>
<tr>
<td>140</td>
<td>3170</td>
<td>3130</td>
<td>3100</td>
<td>3080</td>
<td>3029</td>
<td>3010</td>
<td>2980</td>
<td></td>
</tr>
</tbody>
</table>

The reduction in Colour Temperature suggests that the degradation of the remote phosphor plates has consequences not only on the light extraction efficiency but also on the colour of the emitted light. Colour shifting of light is determined by variation of Chromaticity Coordinate (CIE x,y). The values of x and y of the aged remote phosphors at 140 °C is shown in Table 2. One can see that both x and y decreases by increasing ageing time.

Table 2: (x,y) values of aged samples at 140 °C during high thermal-stress test

<table>
<thead>
<tr>
<th>Ageing time (hrs)</th>
<th>x</th>
<th>y</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.4152</td>
<td>0.3547</td>
</tr>
<tr>
<td>240</td>
<td>0.4149</td>
<td>0.3538</td>
</tr>
<tr>
<td>480</td>
<td>0.4146</td>
<td>0.3528</td>
</tr>
<tr>
<td>720</td>
<td>0.4132</td>
<td>0.3518</td>
</tr>
<tr>
<td>1440</td>
<td>0.4068</td>
<td>0.3461</td>
</tr>
<tr>
<td>2040</td>
<td>0.4053</td>
<td>0.3442</td>
</tr>
<tr>
<td>2880</td>
<td>0.4032</td>
<td>0.3435</td>
</tr>
</tbody>
</table>

4.2. Prediction of time-to-failure at lower temperatures

The real working temperature of LEDs is much lower than the applied temperatures in the tests [13]. Therefore, the kinetics of lumen depreciation to 30% of its initial value by using exponential luminous decay model and Arrhenius equation should be extrapolated to temperatures lower than 100 °C. This can be done using Equation 1 by equating \( \phi \) to 0.7, knowing that \( \alpha \) can be obtained from Equation 2. The values of \( \alpha \), calculated for 40, 60 and 80 °C, are given in Table 3. As is seen the higher the temperature the faster the reaction rate.

Table 3: Reaction rate (\( \alpha \)) for different temperatures

<table>
<thead>
<tr>
<th>Temp (°C)</th>
<th>Remote Phosphor</th>
</tr>
</thead>
<tbody>
<tr>
<td>40</td>
<td>7.95E-06</td>
</tr>
<tr>
<td>60</td>
<td>1.68E-05</td>
</tr>
<tr>
<td>80</td>
<td>3.28E-05</td>
</tr>
<tr>
<td>100</td>
<td>5.93E-05</td>
</tr>
<tr>
<td>120</td>
<td>1.01E-04</td>
</tr>
<tr>
<td>140</td>
<td>1.64E-04</td>
</tr>
</tbody>
</table>

Figure 4 illustrates time-to-failure (70% lumen decay) of remote phosphors, calculated at different temperatures. It is seen that at 40 °C the
light output reduces to 70% of its initial value after 45 khrs.

![Figure 4: Time-to-failure (70% lumen decay) of remote phosphor at different temperatures](image)

5. Discussions:

High temperature stresses can damage the optical properties of the package and of the material used for the encapsulation [1-5]. This can result in a significant reduction in the luminous flux, emitted by the devices. Spectral power distribution (SPD) method is used to study the effect of high temperature stress test on the optical degradation of remote phosphor. The goal was to study the effect of temperature on the lumen depreciation of LED-based products and on their CCTs. A significant change both in the correlated colour temperature (CCT) and in the chromaticity coordinates (CIE x,y) is observed in thermally aged specimens. Worsening of the chromatic properties of the phosphor plates is correlated to the decrease in the luminous flux. One can conclude that the thermal degradation of the remote phosphor plate affects both the efficiency and the colour of the LED products. It is shown that the degradation mechanisms is thermally activated and has activation energy of 0.333 eV (Figure 3). It is clearly seen that the lower the depreciation rate, the better the performance a remote phosphor could have. The results also show that there is a direct relation between the temperature and kinetics of degradation. It is also shown that decreasing the transmissity of PC plates together with the reduction in phosphor efficiency limits the reliability of remote phosphor light sources and there is a colour shift towards yellow. The life time of this kind of remote phosphor is calculated as 45 khrs which is slightly higher than that of other remote phosphors which were studied completely in our previous paper. The reason might be due to the lens material [9].

6. Conclusions:

The accelerated optical degradation of a remote phosphor, under elevated temperature stress, is studied. BPA-PC plates are exposed to temperature in the range of 100 to 140 °C. Exponential luminous decay model and Arrhenius equation are used to predict the lumen depreciation and the lifetime of plastic lens in LED lamps in real service conditions. The following conclusions can be drawn from this study:

- A significant decay both in the phosphor yellow emission and in the blue peak intensity, with blue emission being more influenced.

- During the stress thermal ageing tests a significant change both in the correlated colour temperature (CCT) and in the chromaticity coordinates (CIE x,y) take place.

- The lifetime of the remote phosphor, defined as 30% lumen depreciation at 40 °C, is around 45 khrs for the commercial grades plates.

Acknowledgments

This research was carried out under project number M71.9.10380 in the framework of the Research Program of the Materials innovation institute M2i (www.m2i.nl). The authors would like to thank M2i for funding this project. Authors would also like to acknowledge “TNO innovation for life” company for SPD measurements.

References:


