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A review on discoloration and high accelerated testing of optical materials in LED based-products



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ABSTRACT

Reduction of intensity of light output is one of the most common degradation modes in light-emitting diode (LED) systems. It starts from the failure of the various components in the system, including the chip, the driver, and optical components (i.e. phosphorous layer). The kinetics of degradation in real life applications is relatively slow and in most cases it takes several years to see an obvious deterioration of optical properties. Highly Accelerated Stress Testing (HAST) set-up and a methodology to extrapolate the results to real time applications are therefore needed to test the reliability of LED packages and lens materials. Using HAST concept in LED industry is inevitable due to the necessity of assessing the reliability of new products in a short period of time. This paper aims at briefly clarifying the degradation mechanisms of optical components in LED packages and explaining how they contribute to the depreciation of light output of the LED systems. The concept of HAST and the way the reliability of LED packages can be evaluated will also be discussed.

1. Introduction

Solid state lighting (SSL) technology is known as an innovative invention in the history of the lighting industry. Light emitting diodes (LEDs) are used as a highly efficient source of illumination in solid state lighting systems. During last years many researches have been done about SSL and reliability of LEDs [1-40]. An SSL system is composed of a LED engine with a driver inside a package that enables optical functions, thermal management, protection, and sensing functions [1-2]. LED is a complicated system composed of LED chip, interconnect, substrate and optics. Typical schematics of common LED packaging approaches are shown in Fig. 1.

Efficacy, defined by lumens/watt (lm/W), color rendering index (CRI), and reliability, are main parameters, which make LED a superior power source. Light sources with higher efficacy have obviously higher energy efficiency. Table 1 compares the optical characteristics for common light sources. One can see that the overall properties of LED technology are superior to those of other light sources.

2. Failure mechanisms in LEDs

The failure of LEDs can be attributed to three regions in the system; semiconductors, interconnections and the package [3–7]. Table 2 gives an overview of die-related failures.

Electrical overstress-induced bond wire fracture, electrostatic discharge, and elemental diffusion, leading to catastrophic and unexpected failures of LEDs, are typical interconnect failures of LED packages. Package-related failures are commonly due to the ageing of polymeric materials leading to color change, color shift, optical degradation, and discoloration of the encapsulant/lens materials. Fig. 2 shows some common failures of LED package.

There are many technical challenges for LEDs, when it comes to the lifetime and reliability. Among them, the light extraction efficiency and the light output degradation are main issues, which turn out to be all related to the packaging materials. LEDs have to often operate in different temperatures and environments with high levels of humidity. Ionic contaminants, moisture, radiation, heat, and mechanical stresses can also be extremely damaging to LEDs and can lead to device failures. Almost all microelectronic devices and chips are encapsulated by plastics. LEDs have a longer lifetime, compared to the traditional lighting sources. This obviously means less waste materials and less environmental impacts of the package. Microelectronic components and devices are generally encapsulated by polymers in order to protect the electric circuit from environmental hazards, including ultra violet (UV) light and moisture. A limiting factor in increasing the current standard lifetime of LEDs, aiming for more sustainable and more environmentally friendly products, is the degradation of optical materials. Light Emitting diodes are also encapsulated to avoid thermal and

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Fig. 1. Schematics of a LED package.

 Table 1

 Efficacy, CRI and lifetime of common light sources [3].

	-		
Light Source	Efficacy (lm/W)	CRI	Lifetime (hours)
Incandescent (120 V) Compact fluorescent High-pressure mercury LED	14.4 51 34 130–220	100 80 50 > 80	1000 10,000 24,000 50,000

mechanical stress shock and moisture-induced corrosion and oxidation [8]. Details of package-related failures and the possible solutions are given in Table 3.

Increasing the light extraction efficiency, minimizing the generation of heat, extracting more heat out of the package, and making the package more heat and UV resistant, are all important technical issues to be considered in future developments. In general high refractive index, excellent electrical properties, good chemical resistance, low water absorption (high resistance to moisture), good UV and thermal resistance, good adhesion to package components, and Mechanical strength are essentially needed to improve the performance of the packaging materials and therefore o enhance the lifetime of LEDs.

3. Package-related failures

It is known that the failure of optical components is mainly due to the degradation of the optical component. Meaning that to progress the reliability of LEDs, most efforts have to be done for development of the optical package performance. Some of the most important degradation reasons of optical packages, which result in the ageing of optical materials, are contaminations, interface delamination, and discoloration.

Table 2

Die-related failures in LED light source [3].

In this paper we are focused on discoloration and yellowing of lens materials. Encapsulants, used in LED packages, include plastics, epoxies, or silicones. These materials can discolor to yellow over time. This yellowing will certainly shift color and depreciates optical output of the system. The key reasons of discoloration and yellowing are believed to be constant and long-time radiation of light (blue/UV radiation), working at elevated temperature, and using the phosphor. Increasing the molecular mobility of the polymer as well as the presence of chromophores which is used as an additive in the molecule [10] are main reasons of photodegradation. Exposure time and the amount of radiation are key factors of Photodegradation. The chemistry of degradation processes in polycarbonates, which is one of the most commonly used materials, has been extensively studied over the past few decades [10–12].

4. Chromaticity

"Color maintenance" is analogous to lumen maintenance and is defined as the change in chromaticity of a light source with respect to the chromaticity at the beginning of the lamp's life. It is typically measured as Δxy or as $\Delta u'v'$ in the Commission Internationale de l'Eclairage (CIE) color coordinate systems. The chromaticity coordinates of a source provides a numerical measure of the color of the light. The three most common chromaticity diagrams, with their coordinates, are the CIE 1931 (x, y), the CIE 1960 (u, v), and the CIE 1976 (u', v'). The (x, y) coordinates are the most frequently reported. An example is shown in Fig. 3. Every color is represented by unique (x, y) coordinates. The CIE system is the most commonly used method of characterizing the composition of any color in terms of three primaries [13,14].

The (u', v'u', v') coordinates are related to the (x, y) coordinates by the following equations:

Based on Eqs. (1a) and (1b), the coordinates $\Delta u'v'$, which define the color shift at any two positions (0 and 1), can be calculated using the following formula:

$$u' = \frac{4x}{-2x \times 12y \times 3},\tag{1a}$$

$$v' = \frac{12x}{-2x \times 12y \times 3},$$
(1b)

$$\Delta u'v' = \sqrt{(u_1' - u_0') + (v_1' - v_0')}$$
⁽²⁾

Energy Star specifies that color maintenance must not exceed $\Delta u \dot{v} = 0.007$ on the CIE $u \dot{v}$ diagram, after 6000 h of operation.

"Color consistency" is the variation in chromaticity at the start of a lamp's life among a population of products. For example, a product may be made from LEDs that are binned to fall within three *MacAdam* steps

Failure mechanism	Failure mode	Failure cause	Effect on device
Die cracking	Lumen Degradation, No Light	High ambient temperature High current-induced Joule Heating Poor sawing and grinding process	Thermomechanical Stress
Defect and dislocation generation and movement	Lumen degradation, increase in reverse leakage current, and increase in parasitic series resistance	High current-induced Joule heating	Thermomechanical stress
Dopant unrusion	current,	Junction High current-induced Joule heating High ambient temperature	mermai stress
Electromigration	No light Short circuit	High drive current or high Current density	Electrical verstress



Fig. 2. Schematics of some common failures in LED systems [21].

Table 3

Materials challenges and solutions for packaging high power LEDs [8].

Challenges	Problems	Packaging materials solutions
Light extraction	Refractive index mismatch between LED die and encapsulant	– High refractive index encapslant – Efficient lens/cup design
Thermal yellowing	Thermal degradation of encapsulants induced by high junction temperature between LED die and lead frame	 Modified resins or silicone based encapsulant Low thermal resistance substrate
uv yellowing	Photo degradation of encapsulants induced by UV radiation from LED dies and outdoor radiation	– UV transparent encapsulant
Stress/delamination	Failure of wire-bond and die attach caused by the CTE mismatch among encapsulant, LED die and lead-frame	 Low CTE and modulus encapsulants Excellent adhesion and CTE matching materials between the surfaces

of a target chromaticity. These LEDs have a color consistency of three steps. Color consistency can also be defined in terms of (x, y) or (u', v'). The color consistency of lamps, built from these LEDs, may be worse than three steps because of temperature variations, current variations, or other factors.

The spectral power distribution (SPD) and the yellowing index (*YI*) of the degraded specimens, measured by integrated sphere, are always good measures of the discoloration and yellowing of specimens. The integrated sphere, shown in Fig. 4, is an optical characterization instrument which is used to measuring the light properties. It basically consists of a light source and a detector for optical power measurement. According to the ASTM D1925, the yellowing index (YI) is calculated as follows [50]:

$$YI = \frac{100 (1.28X CIE - 1.6Z CIE)}{Y CIE},$$
(3)

where X and Y are the tristimulus values in (CIE) standard.

Fig. 5 illustrates the color shift $(\Delta u'v')$ of the BPA-PC samples at different stress conditions as well as the yellowing index (YI) of BPA-PC plates as a function of ageing time under thermal and thermal plus bluelight stresses. Clearly during the incubation stage there is no major change in YI, while during the ageing stage the discoloration follows a linear kinetics. Moreover, Fig. 5 b shows that the temperature has a

significant influence on yellowing. It is also obvious that the radiation of the blue light has an important influence on discoloration and yellowing kinetics. The presented results in Fig. 5 are from the ageing test, done on industrially pure BPA-PC, which is used as a lens and as a substrate for phosphor in remote-phosphor LEDs. Details of the test are already published [15]. In commercial samples, the main reason for yellowing is thermal ageing. For this reason, thermal ageing can be used to accelerate degradation reactions in order to study color shift. It is already reported that by increasing the temperature, the discoloration kinetics will be faster [14–15]. (See Fig. 5.)

For examining the effect of harsh environment, it is advisable to use a salt spray set up which simulates the harsh environments with aggressive ionic contaminations. Fig. 6 shows a typical salt spray set-up with an oven. In this such set-up both temperature and oxidizing ionic species are employed to accelerate the discoloration kinetics.

In a series of experiments, performed in our group, the BPA-PC samples were aged in salt atmosphere at 50 °C. Chemical and optical results illustrated that sea side atmosphere have strong effect on the ageing of BPA-PC plates. Fig. 7 shows the transmission of light in aged BPA-PC. It is apparent that by continuing the failure of samples the transmission of light in visible light region decreases.



Fig. 3. The CIE 1931 x,y chromaticity space, also showing the lines of constant correlated color temperature.

5. Reliability

Reliability discusses to the probability that a system, including all components perform its projected function under specified conditions for a stated period of time without ageing in a indicated environment [17]. Modelling of Reliability is related to predict or understand the failure modes of a component or system before the employment. Statistical techniques are used to predict the reliability performance of a component of the system. The LED domain, despite exciting innovations, motivated by technological developments, has still challenges regarding lack of information regarding the failure mechanisms and reliability. The relative low reliability information is an obstacle to the acceptance of LEDs in traditional applications. The lifetime of electronic devices and microchips is shown by lumen maintenance, which is the amount of reduction of light intensity over time. The Alliance for Solid-State Illumination Systems and Technologies (ASSIST) defines LED





b

Fig. 5. Examples of a) Color shift of BPA-PC plate, aged at different temperatures and b) Variation in YI of BPA-PC plates exposed to different stresses [15].

lifetime based on the time to 50% or 70% of light output degradation at room temperature. TM-21 is based on this definition and uses all the raw data, per setting, per LED and per time point. TM-21 is a methodological reference, being established by an Illuminating Engineering Society (IES) technical committee. TM-21 measures the lumen

Fig. 4. Integrated sphere (IS), widely used to measure yellowing index.



Fig. 6. Salt spray test set-up.



Fig. 7. Transmission of light in aged BPA-PC.

maintenance of an LED source, which then be used to measure the predictable light output of the source and eventually can be used to predict the time-to-failure of the system [17]. Table 4 shows the proposed reliability models in TM-21 for different applications and different conditions.

The simplest form of reliability model, which can be applied in many cases for the prediction and evaluation of the LED and phosphor samples, is based on an exponential lumen decay equation, where the Φ can is given as follows:

$$\emptyset(t) = \beta \exp(-\alpha t),\tag{4}$$

Table 4

Lumen maintenance life projections in standard TM-21 [17].

where t is the ageing time in hours; Φ (t) is the normalized luminous light output at time t; α is the reaction rate; β is the constant parameter.

For each separate temperature L_{70} , that is when $\Phi = 0.7$, can then be calculated using averaged normalized luminous flux output.

$$L70 = (-\ln 0.7/\alpha) 1/\beta$$
 (5)

Estimates of (α, β) can be easily obtained by applying the least squares method. Temperature acceleration, within the measured temperatures, is allowed and supposed to follow the Arrhenius equation:

$$\alpha = C \exp\left(\frac{-E_a}{KT}\right) \tag{6}$$

where C is a pre-exponential factor; Ea is the activation energy (in eV).

One of the main challenges in the study of LED reliability is the fact that a single failure mode is governed by multiple failure mechanisms [22]. For instance, the depreciation of the luminous flux, which is used as the main failure criterion [1–6], is affected by the degradations of the different part of the LED (chip, phosphor, and lens). Component degradation is obtained by comparing the optical power of the aged devices before and after ageing. It is observed that for all ageing conditions the degradation of the LED system is mainly governed by the degradation of the optical package [21]. This means that in order to improve the reliability of this family of LEDs, focus has to be put on the development of the optical package reliability. In next part we will focus on acceleration and reliability of lens/phosphor layer of LED system. The phosphor plate is mechanically removed from the system and tested in different conditions.

6. Accelerated and high accelerated stress test (HAST)

A high accelerated stress testing (HAST) experimental set-up was invented and introduced in our previous works to investigate the special effects of radiation of light and the high temperature stress on the kinetics of degradation of phosphor plates used in light source products. HAST is an important improvement and upgrade of our formerly-applied accelerated approach [22], where the thermal stress was the only applied ageing acceleration factor. Applying the elevated temperature stress and light intensity in this newly developed set-up makes the rate of ageing much quicker. The basic part of the HAST contains a blue LED with wavelength of 450 nm and a hot plate. Specimens are located on the hot plate and are directly exposed to light radiation. Absorptive filters are used to avoid the reflection of light by the hot plate. In one series of experiments in our previous work, three different temperatures of 80, 100, and 120 °C are used and samples are degraded up to 3000 h. Phosphor plate were used as experimental samples. Light intensities of

Model	Decay rate	Closed form solution	Comment
1	$\frac{dI}{dt} = k_1$	$I=I^0+k_1(t-t^0)$	
2	$\frac{dI}{dt} = k_1 I$	$I = I^0 \exp\left[k_2(t - t^0)\right]$	
3	$\frac{dI}{dt} = k_1 I + k_2 I$	$I = \left(I^0 + \frac{k_1}{k_2}\right) \exp[k_2(t - t^0)] - \frac{k_1}{k_2}$	Model1 + Model2
4	$\frac{dI}{dt} = \frac{k_3}{t}$	$I = I^0 + k_3 ln\left(\frac{t}{t^0}\right)$	
5	$\frac{dI}{dt} = k_1 + \frac{k_3}{t}$	$I = I^{0} + k_{1}(t - t_{0}) + k_{3}ln\left(\frac{t}{t^{0}}\right)$	Model1 + Model4
6	$\frac{dI}{dt} = K^4 I^2$	$I = \frac{I^0}{I + I^0 k_A (t - t^0)}$	
7	$\frac{dI}{dt} = K_5 \frac{I}{t}$	$I = I^0 \left(t/t^0 \right)^{K_5}$	
8	$\frac{dI}{dt} = k_2 I + K_5 \frac{I}{t}$	$I = (I^{0}) \exp[k_{2}(t - t^{0})] \left(\frac{t}{t^{0}}\right)^{K_{5}}$	Model2 + Model7
9		$I = I^{0} \exp\left[-\frac{(t-t^{0})}{K_{6}}\right]^{K_{7}}$	

825, 3300, and 13,200 W/m² were applied on the sample in the HAST set-up. In HAST set-up, Eyring relationship, given below, is shown to be a more applicable reliability model [17] than Arrhenius model. This has to do with the fact that the intensity of light is also used as an additional factor.

$$R = \gamma_0 (I)^n \exp\left(\frac{-E_a}{KT}\right),\tag{7}$$

where R is the rate of ageing, $\gamma 0$ is constant factor, I is the light intensity, n is the constant factor, Ea is the activation energy (eV) of the ageing, K is the Boltzmann gas constant (eV/K), and T is the absolute temperature (K).

In HAST set-up, the temperatures is ranging from 80 to 120 °C. There is also a temperature increase up to 2, 10, and 20 °C for 825, 3300, and 13,200 W/m^2 intensities respectively. It is important to take this temperature increase into consideration, when it comes to the calculation and prediction of lifetime. The variation in the spectral power distribution (SPD) is a good technique to study the ageing and ageing of optical properties of phosphors plates. Optical properties of photo-aged samples can also be investigated, using an integrated sphere. More details about the experiment and how results can be interpreted are presented in our previous publications [22]. It is shown that by performing accelerated ageing tests in the HAST set-up the ageing of optical part of the system is accelerated up to 25 times for 120 °C and light intensity of 13,200 W/m². Effect of radiation of light on the time-to-failure of the samples is already well explained in our previous publication [28]. As an example, the lifetime of samples, at 40 °C, is around 35 khrs, for the lowest light intensity, which has nearly similar lifetime as thermally-aged samples. The lifetime of the samples with higher energy is reduced to 25 khrs.

7. Conclusions

This paper reviews the research done on the failure mechanisms and reliability of optical materials in LED systems. Different types of failures at the package level are discussed and introduced. Different operational and environmental stresses, including junction temperature, contaminations, humidity, operation temperature, and UV light might cause lumen decay and failure of LEDs. Among different failure modes, thermal degradation is the most severe one. If thermal management is inadequate in the remote phosphor, overheating and damage in the remote phosphor and its substrate can occur. For instance, the adhesives that bind the LED to board may damage due to thermal weakening. Thermal loads result in thermal fatigue of solders in LEDpackage as well. More importantly, thermal ageing results in faster kinetics of yellowing and discoloration. With that said, thermal stress is commonly used for accelerated testing of LEDs. In this paper a new acceleration test method for LED remote phosphors is introduced in which the effect of light intensity (in addition the temperature) on the kinetics of ageing can be examined. The results illustrates that there is a close relation between the light intensity and the loss in conversion efficiency of remote phosphor. In fact, by increasing the light intensity the conversion efficiency of remote phosphor decreases. Among different proposed lumen maintenance models, proposed in TM-21, the generalized Eyring equation is the right choice for the HAST set-up. It is illustrated that the lifetime at 40 °C, is approximately 35 khrs, for the lowest light intensity, is close to the lifetime of aged phosphor under thermal exposure. The lifetime of the phosphors with higher light intensity is predicted to be 25 khrs. Although great effort was put into predicting lumen maintenance, resulting in the LM-80 and TM21 standards, little has been done to address color maintenance. Few (or no) LED-package manufacturers provide warranties on color maintenance. It is an absolute necessity to have a widely accepted method of predicting color maintenance and reliability of LED packages.

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