TU Delft

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Document Version
Final published version

Citation (APA)
Lu, G. (2017). *Solid State Lighting Color Shift*. [Dissertation (TU Delft), Delft University of Technology]. https://doi.org/10.4233/uuid:3a724e0a-4834-458f-af41-60a242052132

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Solid State Lighting Color Shift

PhD Thesis

Guangjun Lu

Solid State Lighting Color Shift

PROEFSCHRIFT

ter verkrijging van de graad van doctor
aan de Technische Universiteit Delft,
op gezag van de Rector Magnificus Prof. Ir. K. C. A. M. Luyben
voorzitter van het College voor Promoties,
in het openbaar te verdedigen

op dinsdag 26 september 2017 om 10:00 uur

door

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Onafhankelijke leden:

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Prof. dr. J. A. Ferreira	Technische Universiteit Delft, reservelid

Guangjun Lu,
Solid State Lighting Color Shift

Ph.D. Thesis, Delft University of Technology
with a summary in Dutch.

Keywords:
Solid State Lighting, Light Emitting Diode, LED Based Luminaire, Color Shift, Color Maintenance, PMMA Degradation, MCPET Degradation, Mid-Power LED Package, Color Shift Acceleration， Color Shift Prediction

ISBN: 97890-6562-4192

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Printed by Delft Academic Press, the Netherlands

To My Parents

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Chapter 1

Introduction

Owing to several advantages such as enhanced energy efficiency and potentially long lifetime, solid-state lighting (SSL) solutions are being investigated to replace traditional incandescent lamps and compact fluorescent lamps in the lighting industry. Increasing electricity prices, emerging concerns about climate change, and desire for energy independence are forcing the global lighting market to shift toward energy-efficient light sources. Europe, North America, Asia, and Australia have enacted bans on the sale of traditional general service incandescent lamps. Significantly, advancements in SSL technology resulting from ongoing R&D and manufacturing enhancement continue to reduce prices, enhance efficacy, and enhance lighting performance, thereby accelerating adoption in many markets regardless of policy directives. As SSL technology has developed, the impacts of SSL will exceed the requirement of energy savings. Moreover, SSL has the potential to exhibit considerable beneficial impacts on the environment, horticulture, livestock production, transportation safety, human health, and productivity. All these benefits can be achieved while saving significant amounts of energy compared to conventional lighting technologies. Recently, many of the leading lighting companies reported that LED lamps and luminaires represent more than 40% of their revenues, including Acuity Brands (55%), Osram (48%), Philips (50%), and Zumtobel (63%) [1–5].

The DOE 2014 SSL Forecast suggested that SSL could account for nearly half of all lighting shipments in the United States (measured in terms of light production capacity in lumen-hours), and approximately 40% of the installed base (in lumen-hours) by 2020 [6]. The United Nations Environmental Programme (UNEP) estimated that the utilization of lighting energy had increased to 2815 terawatt-hours (TWh) in 2010, corresponding to 15% of the total global electricity utility [7]. In this study, it is found that by 2030, the LED technology offers the potential to save 261 TWh annually, a 40% reduction in site electricity consumption, compared to a counter-factual scenario without LEDs. This 261 TWh of savings in site electricity consumption corresponds to 3 quadrillion Btu of primary source energy savings. Furthermore, if the DOE SSL

program objectives are achieved, the total annual energy savings in 2030 would increase to 60%, which is an additional 134 TWh in site electricity or 1.5 quads of primary source energy savings. At an average price of $0.10/kilowatthour, the energy savings would correspond to an annual savings of approximately $40 billion [6].

UNEP estimates that, in the absence of novel policies, 57% of the lighting energy demand in 2030 would be from Asia [7]. Japan led early adoption of LEDs, by prioritizing energy savings following the Fukushima disaster. Other Asian countries, such as China and India, are currently assuming the lead [8]. Japan has led early adoption of LED lighting, encouraged by the high cost of electricity, reduced availability of electricity from nuclear power generators, and rapid technological advances by Japanese LED manufacturers. Fluorescent lighting is currently a dominant competitor, however, the government has proposed to ban the production and importation of fluorescent lamps, beginning in 2020 [9]. The rapid development of the SSL industry in China has been assisted by broad support from the national and regional governments, including subsidies for purchases of manufacturing equipment and lighting products, development of industrial parks, and establishment of standards programs. The national and regional governments have invested considerably in promoting companies at each level of the industry, from epitaxy to luminaire assembly. In addition, compared to Europe and North America, the rapid development of the Chinese economy has produced a greater fraction of the lighting business associated with novel installations, rather than replacement or retrofit of existing equipment. China Solid State Lighting Alliance (CSA) estimated the total production of LED lighting products in 2015 to be 6 billion units [10].

In 2015, VITO, an independent research organization, presented a comprehensive study in Europe, and forecasted that LED lamp sales would increase to 375 million units (22% of all lamps sales), and the penetration of the installed base would increase to approximately 800 million units (7%). It predicts that the average efficacy of all lighting (the installed base of both traditional and SSL sources) would increase from 65 lm/W in 2015 to between 113 and 169 lm/W in 2025 and that the LED penetration will increase to a range between 70% and 86%. The wide range in these estimates shows the importance of further R&D to enhance the LED efficiency and promote faster adoption [11].

Although SSL is still developing, represents only a portion of the installed

base, there is currently a nearly universal acknowledgment that it will eventually become the dominant technology for most lighting applications. SSL is creating an opportunity for an entirely novel lighting system paradigm. Some of the terms that describe the recent innovations in the lighting industry enabled by the emergence of SSL are connected lighting, smart lighting, and adaptive lighting. The convergence of SSL, low-cost sensors, smartphones and apps, and the Internet of Things (IoT) is expected to facilitate novel lighting functionality and an unprecedented exchange of data among lighting and other building systems, the Internet and other devices (e.g., mobile phones). They are widely used in the fields of energy monitoring, smart cities, indoor positioning, broad band communications, and security [5]

LEDs are core components of an SSL lamp or luminaire, and there are predominantly three types of LEDs in the architecture in the general lighting market: the phosphor-converted LED (pc-LED), the hybrid LED (hy-LED), and the red, green, blue, and amber (RGBA) color-mixed LED (cm-LED). The pc-LED is based on a blue LED to pump green and red wavelength optical downconverters (typically phosphors), thereby producing white light. The hy-LED is based on a blue LED, which is used for pumping a green wavelength downconverter; then, the blue and green light is mixed with light from a red LED to produce white light again. The RGBA cm-LED is based on four primary LEDs, namely blue, green, amber, and red LEDs, to produce white light. The pc-LED architecture exhibits a significant room for further enhancement, from the current status of approximately 140 lm/W (warm white) to a potential of 255 lm/W. This potential may be achieved by enhancing the optical light extraction and package efficiency, narrowing the red phosphor emission linewidth, and reducing the blue LED efficiency droop [5].

However, despite the several benefits and additional functionality that can be achieved with SSL, a number of barriers still remain, thereby limiting the adoption of SSL products. These include first cost, reliability, color stability, and some compatibility issues. The major cited failures related to SSL are power/driver components, LED failures (shorts, connections, and board), moisture ingress, corrosion, power quality (surge, noise), lumen depreciation, and color maintenance issue (color shift). Of these, possible lumen maintenance failure and color maintenance are the two essential and important quality and

reliability issues with the exception of some possible catastrophic (abrupt) failures [12]. The lumen maintenance failure of SSL products is generally characterized by L70 life or 70% degradation of the lumen output. However, no failure criterion for color maintenance is still defined particularly in the application field except that the ENERGY STAR® program mandates that Δu'v' at 6000 h of operation does not exceed 0.007 [EPA 2012, 2013] [13], which is perhaps the only industry-wide criterion for color maintenance. It is a reasonable starting point; however, it may not be sufficient to ensure considerably high-quality lighting, particularly because the lifetimes of LED products routinely considerably exceed 6000 h [14]. Poor color maintenance can be a substantial problem in applications where color quality is important, including museum and gallery lighting, architectural facade lighting, retail display lighting, healthcare lighting, hospitality applications, cove and wall wash lighting, and down lighting in commercial and residential applications [15].

1.1 Color Measurements and Coordinate Space

The SSL product measurement is typically performed using an integrating sphere. Typically, the measurement system consists of four parts: (1) light emitting device, (2) light gathering system, (3) light transmitting system, and (4) light analyzing system. The light emitting device provides the AC/DC voltage power connection to the LED product that is producing continuous measureable light. The light gathering system, the integrating sphere redistributes and collects the entire light beam emitting from the SSL product under test. The integrating sphere is an optical component that uniformly scatters the light, which contains a special coating on the surface of the inner sphere. With small exit ports on the sides of sphere, the LED lights can be transmitted through the cosine diffuser, which is a detector, filtering and transferring the distributed light to the cable optical fiber. Then, the light is carried into the Lab sphere spectrometer. The data on the lumen flux and color is collected using software [16].

The three most frequently referenced chromaticity diagrams are the CIE 1931 (x, y), CIE 1960 UCS (u, v), and CIE 1976 UCS (u', v'). Although (x, y) coordinates are most frequently reported, color maintenance is most appropriately documented as the difference or change in (u', v') coordinates, written Δu'v', which is the distance deviating from the reference or original color

point in the CIE 1976 UCS (u', v') diagram, because the (u', v') chromaticity diagram is the most visually uniform (numerical differences are more similar to perceptual differences). The CIE 1976 UCS (u', v') diagram is shown in Figure 1.

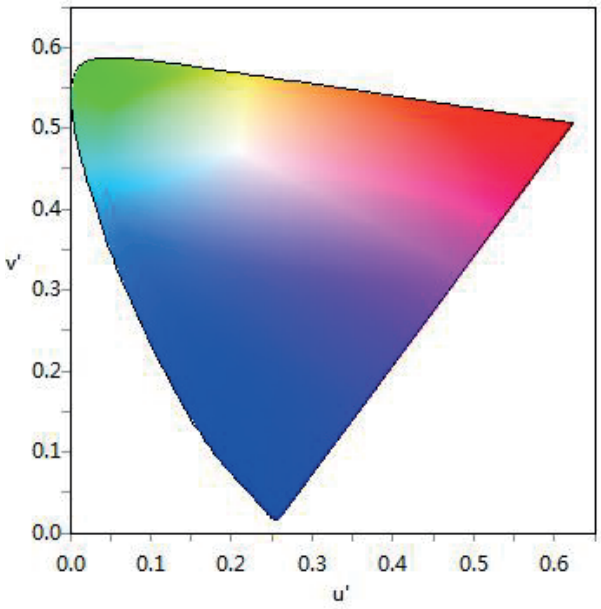


Figure 1 *CIE 1976 UCS（Uniform Chromaticity Scale）(u', v') diagram*

Nonetheless, no existing chromaticity diagram is a perfect representation of human color perception, which itself varies from person to person. For several years, research has been conducted for enhancing the methods used for documenting the color difference. The correlated color temperature (CCT) and Duv are the measures of light source color appearance derived from the CIE 1960 (u, v) chromaticity coordinates, although the concepts are applicable in any chromaticity diagram. In general, the CCT describes a yellow–blue axis, and the Duv describes a red–green axis. Either measure alone cannot accurately describe the exact color appearance of a source. However, when used together, the measures can be considered as a two-dimensional coordinate system. ΔDuv and ΔCCT are insufficient for characterizing the color difference. Only Δu'v' describes the total color difference that might be observed, although it does not describe the direction of the shift. The relationship between CIE 1931 (x, y), CIE

1960 UCS (u, v), and CIE 1976 UCS (u', v') can be described via Equations (1) and (2) [14].

$$u' = u = 4x / (-2x + 12y + 3) \quad (1)$$

$$v' = 1.5v = 9y / (-2x + 12y + 3) \quad (2)$$

Where x and y can be described in the following equations:

$$x = X / (X + Y + Z) \quad (3)$$

$$y = Y / (X + Y + Z) \quad (4)$$

In addition, there is one more parameter z, which can be described as follows:

$$z = Z / (X + Y + Z) \quad (5)$$

When normalized, x + y + z = 1. This is because the two coordinates (x, y) or (u', v') are sufficient for describing the chromaticity in the CIE 1931 (x, y) or CIE 1976 UCS (u', v') chromaticity coordinate system. X, Y, and Z in Equations (3)–(5) can be described in Equations (6)–(8):

$$X = \int_0^\infty I(\lambda)\bar{x}(\lambda)d\lambda \quad (6)$$

$$Y = \int_0^\infty I(\lambda)\bar{y}(\lambda)d\lambda \quad (7)$$

$$Z = \int_0^\infty I(\lambda)\bar{z}(\lambda)d\lambda \quad (8)$$

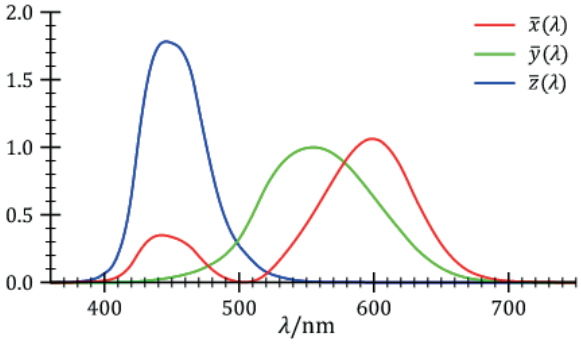


Figure 2 *CIE standard color matching functions*

Where $I(\lambda)$ is the light intensity function on wavelength λ; $\bar{x}(\lambda)$, $\bar{y}(\lambda)$, and $\bar{z}(\lambda)$ are the color matching functions. The CIE standard color matching functions are shown in Figure 2.

1.2 Color Maintenance

A recently appearing system failure mode in LED-based products is color maintenance. Color maintenance problems are insidious, because they are poorly understood and only appear after several hours of operation. The LED Systems Reliability Consortium [17], an industry alliance that is part of the EPA/DOE, has reviewed the studies intended to identify potential failure modes and provide additional understanding of product life. They have reviewed the results of some highly accelerated multivariant tests and other available data for learning the significant failures and the procedures for accelerating those failures. Still further nonpublic information emerges from the experience of the members of the LSRC and enriches the discussions about significant failure mechanisms. Members were asked for the most frequently observed failure modes, and color maintenance was mentioned several times.

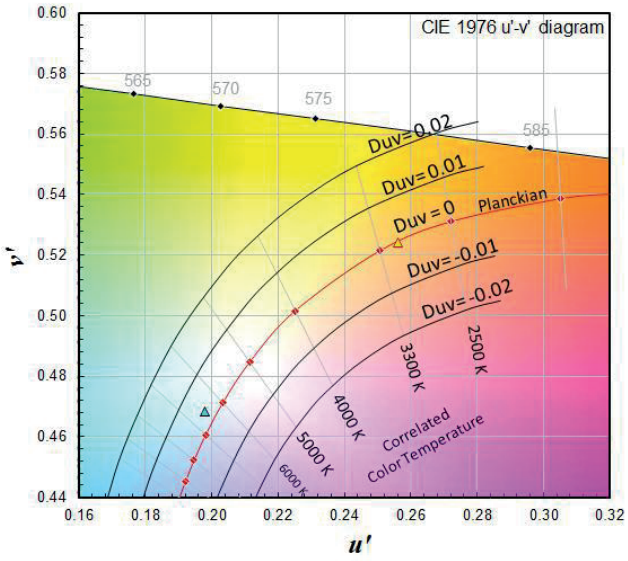


Figure 3 *Duv scale on CIE 1976 (u', v') diagram*

A Delta (uv) (symbol: Duv) value provides information on the distance and direction of a color maintenance issue (color shift from the Planckian locus) (yellowish/greenish or pinkish), whereas the “duv” or another term frequently reveals only the distance and no information on the direction of the shift. With the use of CCT and Duv, the two numbers can provide full information on white light chromaticity of light in an intuitive manner. Duv is defined as the distance from the chromaticity coordinate of the test light source to the closest point on the Planckian locus on the CIE 1976 coordinates, with a plus sign for above and a minus sign for below the Planckian locus [ANSI 2008]. The Duv scale on the CIE 1976 (u', v') diagram is shown in Figure 3 [18-23].

1.3 Testing SSL Products

This paragraph describes the reliability tests currently used for SSL products. Preventing and/or discovering failure modes at the earliest possible integration level will enable considerable cost savings. The reliable products start with understanding the physics of failure by using accelerated test approaches such as (highly) accelerated life testing. A classical reliability approach is to use the results from these tests, verified by failure analysis, for obtaining the conservative bounds from the failure models and predicting the failure rates on a system level. The lighting industry is governed by test standards, in Europe predominantly emerging from the International Electrotechnical Commission (IEC) [24] and the International Organization for Standardization (ISO) [25]. Most of the international companies participate in these standardization bodies to maintain a tap on technology development and to avoid the rules written by their competitors to comply with in the future. No matter how big or small your company is, your voice counts equally. You should know at the design stage what differs where, so as to avoid multiple store keeping units (SKUs), duplication and waste of money and time. Your technology will be globally recognized and you can still protect your IP. Standardization bodies have put in place high-level technology watch mechanisms that enable it to develop the right deliverables exactly when you require them. By prioritizing standardization and conformity assessment operation and positioning novel structures, tools, and processes as well as cooperating with all relevant organizations, the bodies help to sell your products rapidly to more markets. Being one of the activities, these

standardization bodies cover the so-called environmental effects in accelerated tests. The conditions and durations may differ from product to product, or even from country to country. One of the mostly used standards is EN 60068: Environmental testing [26]. EN 60068 provides a standard procedure for determining the ability of a specimen to withstand specified severities of nonrepetitive or repetitive loading. The purpose of this test is to reveal mechanical weakness and/or degradation in specified performances, or accumulated damage or degradation caused by these loads. In conjunction with the relevant specification, this may be used in some cases to determine the structural integrity of specimens or as a method of quality control. These tests are primarily intended for unpackaged specimens and for items in their transport case when the latter may be considered to be part of the specimen. There are quite some tests described under this standard. A few examples are provided in the following:

- EN 60068-2-1 Cold temperature
- EN 60068-2-2 Dry heat
- EN 60068-2-11 Salt mist
- EN 60068-2-13 Low air pressure
- EN 60068-2-14 Change of temperature
- EN 60068-2-17 Sealing
- EN 60068-2-18 Water
- EN 60068-2-21 Robustness of terminations and integral mounting devices
- EN 60068-2-27 Shock
- EN 60068-2-29 Bump
- EN 60068-2-30 Damp heat
- EN 60068-2-31 Drop and tumble
- EN 60068-2-32 Free fall

Table 1 lists the test requirements as specified by the standard. This list can be viewed as the test requirements for the SSL products. Either the complete product or its subsystems should be submitted to either one of these tests.

Some standards have been generated and widely used for the SSL products in the measurement of lumen, lumen maintenance, color and color maintenance from the LED package level to the lamp or luminaire level, as shown in Table 2. IEC TS 62861:2017 provided guidelines for principal component reliability testing for LED light sources and LED Luminaires [27]. However, in terms of prediction, only lumen maintenance exhibits standards both in LED package level and in luminaire level, and no such standard is available for color maintenance prediction.

Particularly, IES LM-80-08 details the procedures for measuring chromaticity for LED packages over time; however, there is no method analogous to IES TM-21-11-for predicting future lumen maintenance from the measured data-for predicting color maintenance over time [28-29]. Similarly, there is a method TM-28 on lumen maintenance prediction for LED-based luminaire level products. However, no such method is available for color maintenance prediction although LM-84 has been generated and a committee is investigating to provide a method for measuring both lumen and color maintenance.

Table 1 Selection of IEC tests used for lighting products

Environmental effects	Description	Test standard	Typical environmental conditions	Typical duration
Climatic	Cold	EN 60068-2-1	-55 °C	
	Heat	EN 60068-2-2	100 °C	1000hrs
	Heat	EN 60068-2-2	125 °C	1000hrs
	Heat	EN 60068-2-2	150 °C	1000hrs
	Thermal Change	EN 60068-2-14	-40 °C to + 125 °C	
	Damp heat (constant, cyclic)	EN 60068-2-3 EN 60068-2-38 EN 60068-2-78	85 °C / 85% R.H.	1000hrs
	Simulated solar and UV radiation	EN 60068-2-5		1000 hrs
	High or low pressure	EN 60068-2-13 EN 60068-2-41		
	Degree of protection: sand and dust	EN 60529 EN 60068-2-68		
	water, spray, rain	EN 60529, DIN 40050		
Chemical	Corrosion tests, humidity resistance tests	EN ISO 9227, ISO 6988, EN 60068-2-52, EN ISO 6270-2		

	H2S, SO2 resistance tests	EN/JIS C 60068-2-43, ASTM B117, ASTM G85	H2S 15 ppm 40C/80%
Mechanical	vibration (sine, random, sine on random, …)	EN 60068-2-6 EN 60068-2-64 EN 61373	
	shocks and impacts	EN 60068-2-27	
	combined tests	IEC 68-2-50,	
	heat/cold/humidity - vibration/shocks	IEC 68-2-51, IEC 68-2-53	
			-100 °C to + 200 °C,
	Pressure cooker test		up to 100 gRMS
Robustness	HALT Testing		120 °C, 85% R.H.

Table 2 Selection of IES Standards used for SSL products

ID	Name	Purpose
LM-79	Electrical and Photometric Measurements of Solid-State Lighting Products	To provide procedures for reproducible measurements of photometry, color and electrical characteristics of SSL products
LM-80	Measuring Lumen Maintenance of LED Light Sources	To provide methods of the measurement of lumen maintenance, color of LED packages, arrays and modules
TM-21	Projecting Long Term Lumen Maintenance of LED Light Sources	To provide a calculation tool to interpret the data collected from LM-80 testing; to provide users with lumen maintenance life (e.g., L70) projection, or to predict estimated lumen output values at a given time duration; to interpolate lumen maintenance behaviors for the in-situ temperature (different from testing temperature)
LM-82	Characterization of LED Light Engines and LED Lamps for Electrical and Photometric Properties as a Function of Temperature	To describe the procedures in performing reproducible measurements of LED light engines and integrated LED lamps, at any given temperature for the performance characteristics (total luminous flux, electrical power, etc.)
LM-84	IES Approved Method for Measuring Lumen and Color Maintenance LED Lamps, Lighting engines, and Luminaires	To provide the method for measurement of lumen and color maintenance of LED lamps, light engines, and LED luminaires.
TM-28	Projecting long-term lumen maintenance for LED lamps and luminaires	To develop a LED lamp and luminaire level counterpart to TM-21 using the new LM-80 and LM-84 testing data for projecting long-term lumen maintenance.

1.4 Origins of Color Maintenance Degradation

Any source of lumen depreciation is likely to be a source of color maintenance as well, because the light output degradation is not generally completely uniform across the entire spectrum. Different degradations in different parts of the spectrum will inevitably lead to color changes, although the

amount of color maintenance may be small. The most common origins of color maintenance in LED-based products are described in Table 3. This table lists various mechanisms within an LED-based product that may affect the color of the light emitted by that LED over time.

Table 3: Origins of color maintenance

Root cause	Examples
Material Degradation	• Degradation of direct optical path from LED die to air • Degradation of reflective surfaces within the LED component • Degradation of the optical materials with the system, be it MCPET, white solder resist, Poly Carbonate or PMMA.
External Contaminants	• Contaminations in the direct optical path such as browning of the optical path due to VOCs or residual flux after reflow on the exterior of the LED package. • Change in the reflective surface properties of materials within the LED component, including, for example, tarnishing of silver. • Carbonization due to lack of oxygen. • Sedation of particles onto any optical surface, for example, onto the silicones.
Interface Delamination	• Separation between different material interfaces such as substrate and optical path materials. • Material cracking, for example, in the MCPET reflector due to brittleness.

Details on the possible mechanism are described in the next paragraph.

1.5 Color Maintenance Mechanisms and Models

Unlike traditional lighting products, the color maintenance mechanism of LED lighting is complex owing to its considerably more comprehensive structure, generally composed of LED die, phosphor, silicone, reflector, diffusers, and so on, all of which may contribute to the color maintenance during

operation and each individual component exhibits its own degradation mechanism [12].

The factors impacting color point stability in LEDs include aging-induced changes in the phosphor, emitter, and encapsulate materials. Emitters can exhibit decreases in radiant flux over time; phosphors can experience decreases in quantum efficiency or shifts in emission spectrum owing to oxidation; and encapsulates can exhibit cracking, oxidation and yellowing, or changes in the index of refraction. Higher temperatures will accelerate these degradation mechanisms leading to greater color maintenance; however, the magnitude of the color maintenance as a function of temperature will vary with packaging materials and manufacturing processes. The resulting direction of color maintenance depends on the dominant degradation mechanisms occurring in the package, which in turn depend on the materials of the packages and the methods of construction [30-31].

Some studies were performed on some failure mechanisms of package level color maintenance, such as the color maintenance to the blue end of the spectrum caused by the settling of phosphor for the combination of “blue” LED and “yellow” phosphor, the color maintenance caused by the discoloration of the plastic (PPA) or polycarbonate (PC) used in the package, and the color maintenance toward yellow end resulting from the mean free path of the blue photons through the increase in phosphor caused by delamination, and so on [32–41].

Meneghini and Zanoni et al. investigated InGaN/GaN-based high brightness LEDs with high temperature or dc or pulsed stress. The results revealed that thermal treatment can exacerbate the chromatic properties of the devices: a decrease of the yellow emission intensity was detected after stress, with subsequent shift of the output of the devices toward bluish light. Further analysis indicates the carbonization of white plastics implying a reduction of package reflective properties and the darkening of the top-side contact layer. The darkening of the plastic reflector reduces the overall volume of the phosphors that effectively plays a role in wavelength conversion, thereby implying the changes of the spectral properties of the LEDs and possibly that stress can also exacerbate the conversion efficiency of the phosphorous material [42–47].

Huang et al. studied the color maintenance caused by the yellowing of the package encapsulated for mid-power white-light LED packages in the outdoor illumination applications with high humidity and high temperature (WHTOL) [48]. Mehr et al. experimentally investigated the color maintenance of remote phosphor plates fabricated from Bisphenol-A polycarbonate (BPA-PC) [49–52].

Davis et al. examined the chromaticity shift modes of the PAR38 lamps with four types of built-in LED packages. The results revealed that the chromaticity shift is dominated by the characteristics of the LED, because the optical materials changed slightly in most samples. The four primary potential chromaticity-shift directions of a light source are blue shift, yellow shift, green shift, and red shift. A blue shift may result from an increase in blue emission or a decrease in yellow emission with chromaticity decrease in both u' and v'. Such shift can be attributed to loss of phosphor quantum efficiency owing to chemical change or temperature effects, oxidation of the molding compound in the PLCC, operating the phosphor above the saturation flux level settling and precipitation of the phosphor, and/or top-to-bottom fractures of the binder in the phosphor-binder layer, thereby resulting in blue photons bypassing the phosphor layer. The yellow shift may result from an increase in yellow emission or a decrease in blue emission with chromaticity increase in both u' and v'. The potential causes are the increase in phosphor quantum efficiency owing to chemical changes or temperature decreases, cracking or delamination of the phosphor-binder layer, discoloration/oxidation of the lenses, and/or discoloration of the reflector. The green shift occurs when the chromaticity moves away from the blue–yellow line and proceeds toward the green direction with a chromaticity decrease in u' and a minimal change, and possibly an increase in some cases, in v'. A green shift indicates a change in the emission properties of either the blue or yellow emitter. The green shift could be caused by the oxidation of phosphors. The red shift occurs when the chromaticity moves away from the blue–yellow line and proceeds toward the red direction with a chromaticity increase in u' and a minimal change in v'. A red shift indicates a change in the emission properties of either the blue or the yellow emitter. A shift in the emission properties of direct red emitter could also induce such shift [53–57].

Lall et al. [16] used the exponential model, as shown in Equation (9), for predicting the CCT (correlated color temperature) and lumen maintenance for both LED lamps and LEDs inside the LED lamps subjected to 85°C/85%RH test.

$$\Phi = \beta e^{\alpha t} \tag{9}$$

Where β is the pre-decay factor, α is the decay rate, t is the test time, and Φ is the CCT or lumen maintenance depending on the decay rate being calculated. The decay rate is a function of temperature and represented by Equation (10), which is also known as the Arrhenius equation.

$$\alpha = Ae^{-\frac{Ea}{K_b T}} \tag{10}$$

Where T is the temperature in kelvin, K_b is the Boltzmann constant, and Ea is the activation energy. The method of least squares (LS) was used for computing the decay rate for both CCT and lumen maintenance.

Mehr et al. generalized a Eyring equation-reliability model, as shown in Equation (11), for predicting the lumen maintenance and the CCT for the remote phosphor plates subjected to a HAST test, which is an accelerated combination of light intensity and temperature.

$$R = \gamma_0 (I)^n e^{-\frac{Ea}{K_b T}} \tag{11}$$

Where R is the reaction rate, γ_0 is the pre-exponential factor, Ⅰ is the intensity of the blue light, n is the constant factor, Ea is the activation energy, Kb is the Boltzmann constant, and T is the absolute temperature [58].

In addition, Koh et al. demonstrated that a linear model could be applicable for the color maintenance of LED devices by analyzing both the experimental and simulation data [59]. Huang used such a linear model to predict the color maintenance of mid-power white-light LED packages, and the decay rate constant was estimated using the modified Wiener process. The data of early degradation was excluded [37].

1.6 Challenges and Objectives

As mentioned previously, color maintenance problems are insidious, because they are poorly understood and only appear after several hours of operation. As such, color maintenance acceleration models are required for verifying the prediction method particularly for LED-based luminaires. The reliability of such calculations is presently a concern, although the IES has initiated a committee

that will be charged with developing an approved procedure. This investigation is targeted toward LED packages, for which the exact operating characteristics are more easily controlled. Considerably similar to lumen maintenance and reliability, extending the component-level data to complete LED lamps and luminaires can be difficult and challenging [14]. A lack of standard procedures for predicting color stability performance has contributed to user uncertainty—potentially limiting adoption and making it more challenging for manufacturers to provide warranty coverage for color maintenance [15].

Challenges with LED lamp or luminaire level color maintenance predictions are as follows:

- Color maintenance mechanisms caused by LED packages under different aging conditions
  Color varies with the change of spectral properties (spectral power distribution), which could be ascribed to different mechanisms such as degradation of active layers, decay of the conversion efficiency of phosphors, modification of the reflective properties of packages, and degradation of the transmittance of lens if present. High current, high temperature or humidity stress exhibits different impacts on color maintenance mechanisms.
- Color maintenance prediction caused by LED packages
  Even submitted to the same environmental stress, the color maintenance could be different with different LED packages. Moreover, different parts in LED packages exhibit different degradation kinetics.
- Color maintenance mechanisms caused by diffusers
  Some publications are available on the degradation of some diffuser materials; however, publications on the related color maintenance mechanisms are scarce.
- Color maintenance effects and prediction caused by diffusers
  The transmittance changes with the degradation of diffusion, which successively alters the SPD of the lighting exit; however, the magnitude and kinetics are unknown and are difficult to predict.
- Color maintenance mechanisms caused by reflectors
  Different reflection materials may exhibit different mechanisms. Moreover, considerably scarce publication was reported on this topic.
- Color maintenance effects and prediction caused by reflectors

The reflectance changes with the degradation of reflectors, which successively changes the color of lighting owing to the SPD change; however, the kinetics and magnitude are still unknown

- Investigation method for color maintenance quantification caused by each individual component
  Each individual component exhibits its own degradation kinetics and effects on the color maintenance; however, it is difficult to investigate.
- Acceleration method or approach for luminaire level color maintenance
  Each individual component exhibits its own degradation kinetics, and it is a challenge to develop a method or approach for acceleration.
- Prediction for luminaire level color maintenance
  Owing to its high complexity and lack of methods and approaches, prediction of color maintenance still remains a challenge to researchers.

The **primary objective** of this thesis is the following:

Build an acceleration method and accompanying acceleration model that is able to accurately predict luminaire level color maintenance over the complete lifetime of the product.

The objectives related to this primary objective are as follows:

- Color maintenance mechanisms investigation for predominant diffuser materials: BPA-PC and PMMA
  BPA-PC and PMMA are widely used as the diffuser material for solid state lighting, and considerably available color maintenance mechanisms were not reported.
- Color maintenance mechanisms investigation for predominantly reflective materials: MCPET and MCPOLYCA
  MCPET and MCPOLYCA are two types of emerging reflective materials used for LED-based luminaires that exhibit high reflectance.
- Color maintenance investigation for mid-power LED packages
  Mid-power LED packages, whose electrical power rating ranges from 0.2 to 0.5 W, are currently widely used in many indoor illumination applications and instead of other high cost devices owing to many advantages such as cost-effectiveness and ease of installation for the simpler design.
- Investigation method for color maintenance quantification caused by each individual component

It is essential to develop a method for investigating the magnitude of color maintenance caused by each individual component.

- Acceleration method or approach for luminaire level color maintenance
  Each individual component exhibits its own degradation kinetics, and the objective is to develop a method or approach for acceleration.
- Prediction for luminaire level color maintenance
  The acceleration method will be used for predicting the luminaire level color maintenance.

1.7 Outline of the Thesis

This thesis is written on the basis of journal publications (or under review) and/or contributions to conference papers with some small overlap in few chapters. Each chapter can be read independently. The structure of this thesis is organized as follows:

In Chapter 2, color maintenance mechanisms for LED secondary optical designs were investigated and comparisons between two widely used diffusers, BPA-PC and PMMA, were performed.

In Chapter 3, color maintenance and mechanism were further investigated using more severe aging conditions compared with those used in Chapter 2 on the PMMA diffuser used in LED-based luminaires, and some helpful conclusions were obtained.

In Chapter 4, degradation of microcellular PET reflective materials used in LED-based products was studied. Both the lumen maintenance and color maintenance were investigated. Failure modes and degradation mechanisms were also investigated.

In Chapter 5, Color maintenances of widely used mid-power white-light LED packages under different aging conditions were investigated. The type of mid-power LED package investigated is also widely used in the downlight luminaire, which is presented in Chapters 6 and 7.

In Chapter 6, a novel approach is proposed for color maintenance investigation on LED-based luminaires. Using this approach, color maintenance contribution of each individual component (diffuser, reflector, housing, and LED package) could also be calculated besides luminaire level color maintenance.

In Chapter 7, based on the approach proposed in Chapter 6 and some experimental results, LED-based luminaire color maintenance acceleration method and prediction approach are illustrated and proposed. Prediction results are also presented.

In Chapter 8, guidelines and design rules based on the results and findings in the previous investigation in Chapter 7 are provided, and these guidelines will be helpful in designing or manufacturing LED-based luminaires.

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Chapter 2

Color Shift Investigations for LED Secondary Optical Designs：Comparison between BPA-PC and PMMA[1]

In this chapter, broadly used commercial duffuser materials (BPA-PC and PMMA) are experimentally investigated on the color shift effects during aging. Besides this, color shift mechanisms of degradation of transmittance are also studied. Results revealed: 1) Inconsistent degradation of wavelength–dependent transmittance induces the decrease of the blue/yellow light intensity ratio and thus gives rise to the color shift toward the yellow field, which is the color shift mechanism of BPA-PC; 2) Even for the non-aged BPA-PC, the transmittance varies with wavelength in the visible light field due to the chemistry of the material，which caused the change of intensify ratio of blue light to yellow light in the SPD， leading to color change in perception; 3) Oxidation plays a key role in the degradation of transmittance at around the peak wavelength of the blue light field, which is in correlation with the discoloration of thermally-aged BPA-PC materials. By contrast, for the PMMA specimen aged up to 3000 hours, oxidation was neither occurred at 85deg.C, nor with additional exposure to blue light, nor even with the additional humidity of 85%RH.

[1] This Chapter is derived from the publication: Guangjun Lu, M. Yazdan Mehr, W.D. van Driel, Xuejun Fan, Jiajie Fan, K.M.B. Jansen, G.Q. Zhang， Color shift investigations for LED secondary optical designs: Comparison between BPA-PC and PMMA, Opt. Mater. 45(2015) 37-41

2.1 Introduction

LED lighting products are dominating the general lighting market due to continuous efforts on cost reduction, quality improvement and incentives from governments. Luminous flux maintenance and color stability are two important factors to evaluate the long term lighting performance, which are also concerns of potential consumers and limit the widespread in some degree. Many research efforts are put on the lumen maintenance over time and even some standards are published to enable an extrapolation, or even for acceleration. For example, IES-LM-80 combined with TM-21 to measure and project lumen maintenance of LED light sources, and CSA-020 used for accelerating lumen depreciation test for LED-based lighting products [1-3]. However, research activities and achievements on the color stability have lagged behind. With increased adoption and accumulation of hours of use, awareness is growing that color shift is an issue for some products especially in the important application fields, such as museums, patient examination rooms, urban scene illumination occasions, offices, street lighting and so on [4-5].

Previous studies on color shift are performed mainly at LED die or package level. For example, Chhajed et al. reported the influence of junction temperature on chromaticity of trichromatic white-light sources and their experiments demonstrated that the color shift phenomenon was observed as the junction temperature increased from 20deg.C to 80deg.C [6]; Yanagisawa et al. reported the color quality impact of long term accelerated current operation of white light–emitting diodes under a high-humidity environment and found the change in the emission spectrum, which could induce color shift, was slight [7]; Trevisanello et al. reported color shift of high brightness LED both in the electrical stress and in the thermal stress and their analysis on the package (before and after stress) indicated high temperature levels induced the carbonization of the plastic package of the devices which was thought to be responsible for the decrease of the yellow emission and gave rise to color shift [8].

However, unlike traditional lighting products, color shift mechanism of LED lighting is complex, because it relates to the degradation of many components, e.g. LED epitaxial layout, phosphor, lens, reflector, diffuser, electrical driver and

so on, and probably each individual component has its own degradation mechanism [9]. LED lens and diffuser are important parts for the secondary optical design to optimize the light extraction process and increase the light use efficiency. Previous studies have demonstrated that thermal ageing stress and UV or blue light irradiation induced defects and the browning or yellowing of the encapsulating lens and made the optical power decrease gradually [10-11], which could potentially induce the output spectral shape change and chromaticity shift. However, further investigations on color shift mechanisms are not publically available, i.e. correlation between color shift and degradation of optical parameters of materials and yellowing. BPA-PC (Biphenyl A polycarbonate) and PMMA (Poly -methyl methacrylate) are broadly used as lens or diffuser materials for indoor lighting application due to comparatively low cost and mature manufacturing process. This paper will experimentally investigate the color shift effects of aging stress and color shift mechanism based on those two materials. Results of this investigation and conclusions drawn in this paper will help guide high quality LED luminaire designs and applications in terms of long-term color stability.

The remainder of this paper is organized as follows: Section 2 presents the materials and methods for the experiment set-up and testing. Section 3 provides results and discussions on the designed experiment. Finally, concluding remarks are presented in Section 4.

2.2 Materials and Methods

Same thickness (3mm) of round commercial BPA-PC plates with a diameter of 50mm and rectangular commercial PMMA plates with a dimension of 30mm×50mm were submitted to the thermal aging test in the isothermal oven up to 3000 hours with a setting of 85deg.C. Molecular structures of BPA-PC and PMMA monomer are presented in Figure1 and Figure 2 respectively. Some special UV protection coating was formed on the surface of the BPA-PC plate, while for the PMMA, there is a pre-existing surface protective tape attached to its surface and before submitted to the aging stress, the protective tape was removed. Additionally, blue light irradiation and humidity reliability test items were respectively done for the same type of PMMA specimen in order to

investigate the color shift effects of different stress. In the blue light irradiation test, the specimens were placed inside the oven with a setting of 85deg.C and exposed to the blue light through the oven glass window generated by the blue light LEDs outside the oven, and the distance between blue light source and the specimens is 40cm and the blue light intensity that the specimen received is around 40k lux, the peak wavelength of the blue light source is 450nm; humidity reliability test item was done in the isothermal oven up to 3000 hours with a setting of temperature of 85deg.C and relative humility of 85% RH.

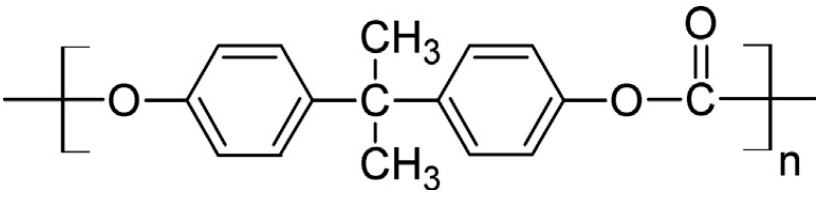


Figure 1: *Molecular structure of BPA-PC*

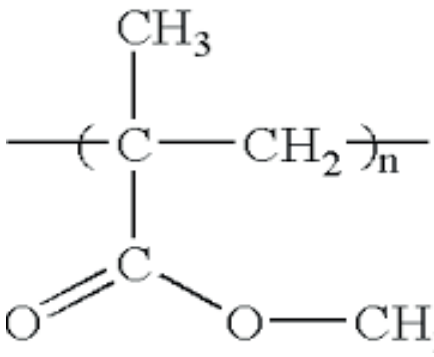


Figure 2: *Molecular structure of PMMA*

Spectral power distribution (SPD) and chromaticity coordinate data of specimens involved LED Package (as light source) and the LED package itself were measured by an integrated sphere with a diameter of 1m. The specimens investigated were mounted very close (~5mm) to the LED package in order to avoid the impact of specimen shape on the measurement result. Infrared spectra of specimens with different ageing conditions were measured using a Perkin–

Elmer Spectrum 100 series spectrometer in the attenuated total reflection (ATR) mode at a resolution of 1 cm^{-1}. Transmittance spectra of BPA-PC and PMMA in the range of visible light wavelength field of 400–800 nm, were measured and recorded by the Lambda 950 spectrophotometer (PerkinElmer 950).

2.3 Results and Discussions

After 3000hours of thermal aging at 85deg.C in the oven, the color of BPA-PC tends to be yellowing. By contrast, discoloration for the PMMA specimens was neither observed at thermal aging of 85deg.C, nor with additional blue light irradiation, nor with humidity of 85%RH. Pictures of the specimens before and after aging stress are shown in Figure 3.

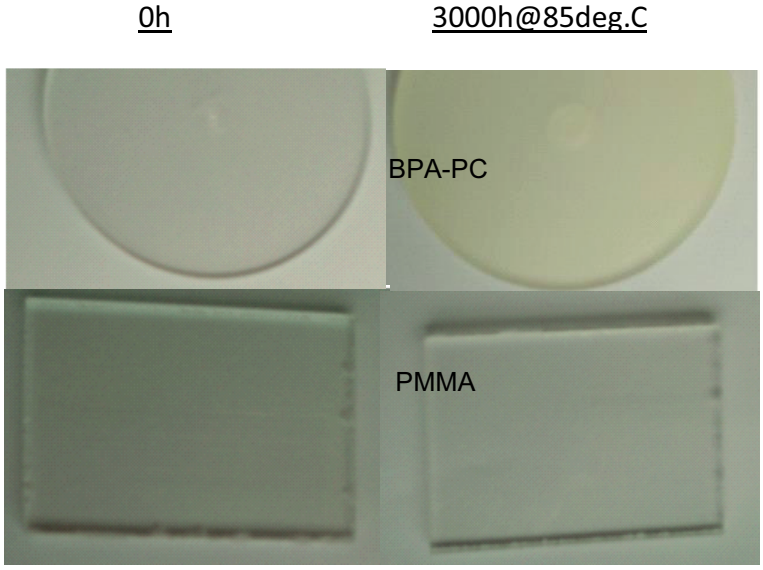
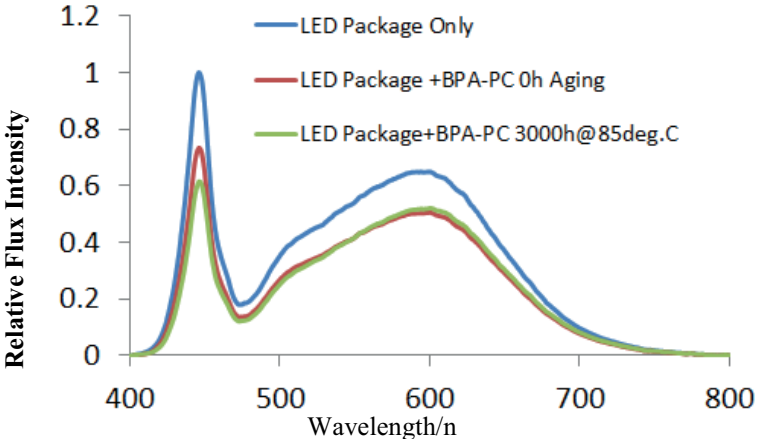


Figure 3: *Specimens before and after aging*

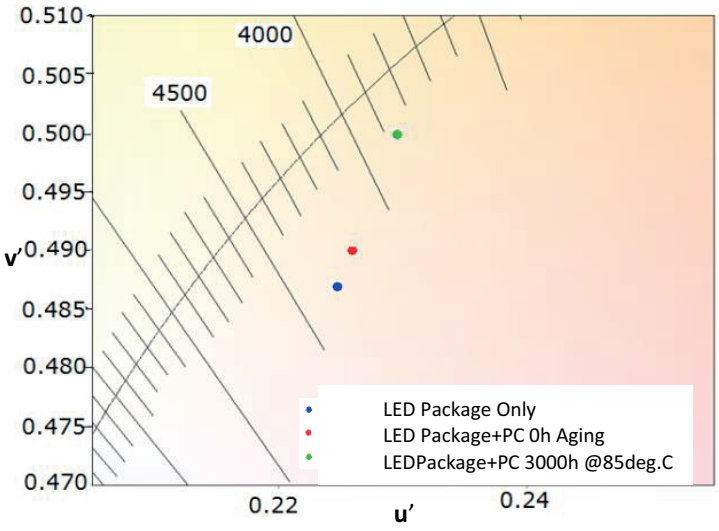
2.3.1 Chromaticity and SPD Results

2.3.1.1 Results of BPA-PC

A commercially available mid-power phosphor-converted LED package (type 5630 [12]), not aged yet, was used as a light source in the investigation of chromaticity change of BPA-PC specimen. SPDs and chromaticity properties of the light source, the light source mounted with non-aged BPA-PC specimen, and the light source mounted with aged BPA-PC specimens were measured and recorded respectively by an integrated sphere under 2pi mode. Results were shown in Figure 4 a) and b) respectively.



a) SPDs of BPA-PC



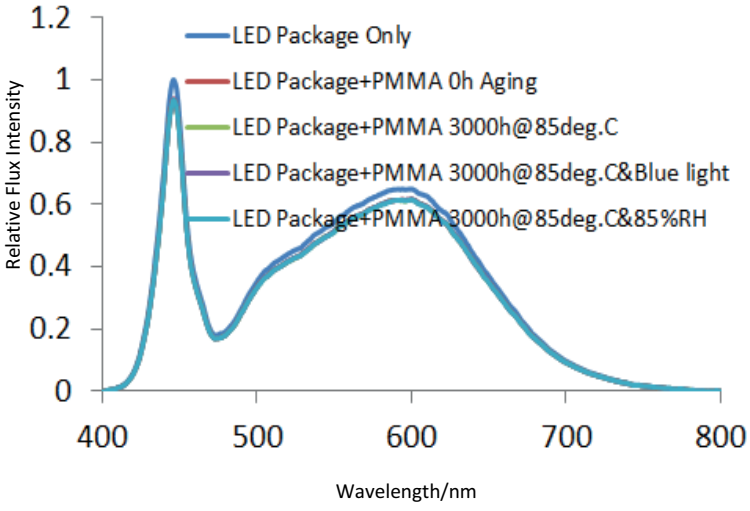
b) Chromaticity of BPA-PC

Figure 4: *SPDs and Chromaticity of BPA-PC*

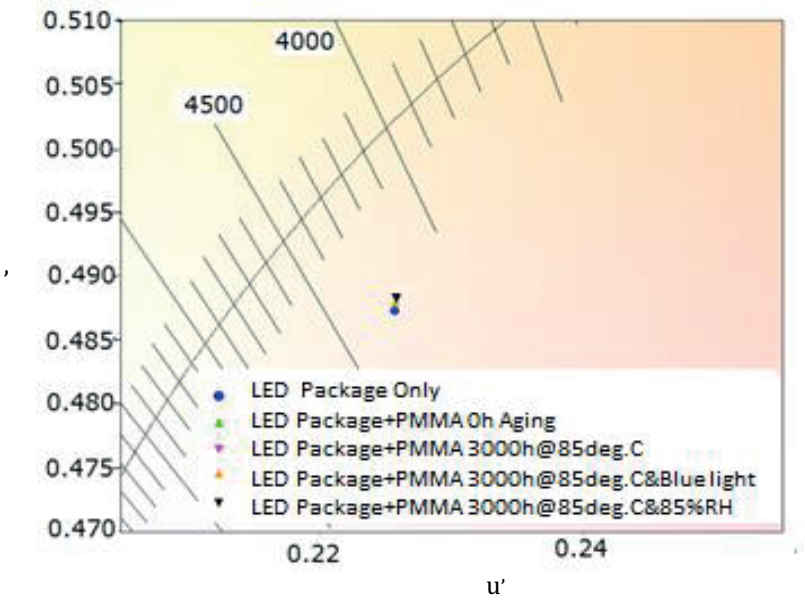
As shown in Figure 4a), after BPA-PC mounted, thermally aged or not, the relative luminous intensity decreased both in the wavelength range of blue light (around 450nm) and in the yellow light range (around 600nm). Compared with the non-aged PC, the relative luminous intensity of thermally-aged PC decreased obviously at around the peak wavelength of blue light, while remained stable in the range of the yellow light. Accordingly, the chromaticity shifted a lot toward yellow area as shown in Figure 4b).

2.3.1.2 Results of PMMA

Similarly, SPD and chromaticity of PMMA specimens under different aging conditions were investigated and shown in Figure 5a) and 5b) respectively. No significant difference was observed. However, as shown in Figure 5a), slight intensity decrease was found both at around the peak wavelength of blue light and in the range of yellow light field after PMMA (aged or non-aged) was mounted.



a) SPDs of PMMA

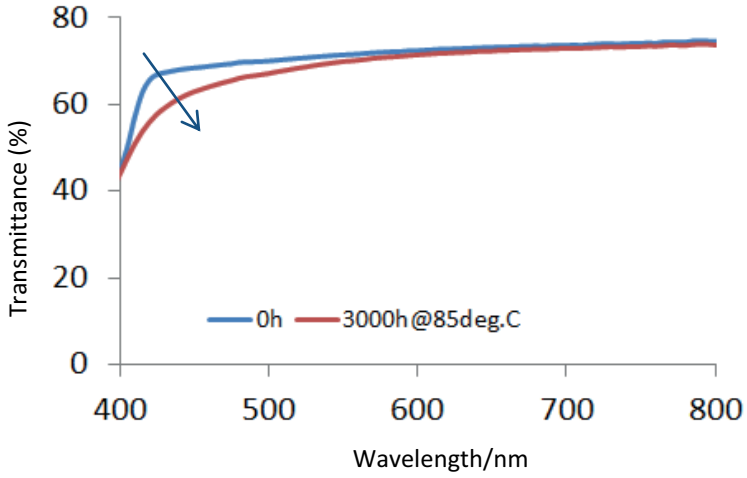


b) Chromaticity of PMMA

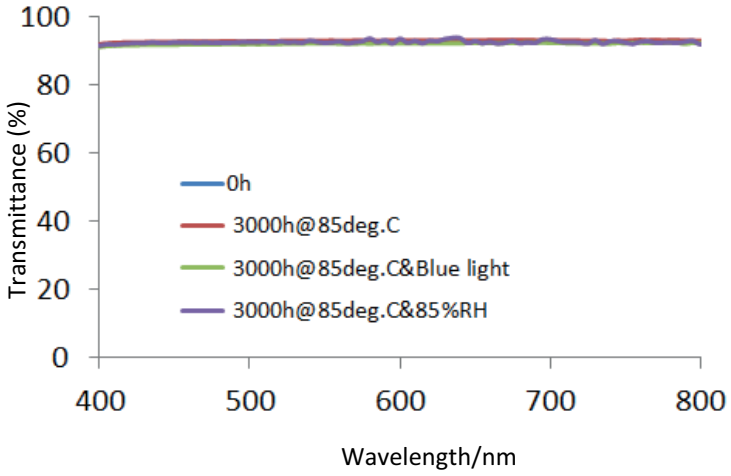
Figure 5: *SPDs and Chromaticity of PMMA*

2.3.2 Transmittance measurements

To investigate the mechanisms of above phenomena, the transmittance spectra of the BPA-PC and PMMA specimens, ranging from 400 to 800 nm with a step of 5nm, are measured and recorded by the spectrophotometer Lambda 950. Results are shown in Figure 6.



a) Transmittance Spectra of BPA-PC



b) Transmittance Spectra of PMMA

***Figure 6**: Transmittance spectra of PMMA and PC*

As shown in Figure 6a), the transmittance of BPA-PC at around the peak wavelength of blue light (around 450nm) decreased obviously after 3000 hours of thermal aging, which could definitely induce the decrease of the luminous intensity of blue light in the SPD (as shown in Figure 4a)) and result in the decrease of intensity ratio of blue light to yellow light in the combination of white light, leading to the color shift toward the yellow area. In addition, even for the non-aged BPA-PC, the transmittance varies with wavelength. The transmittance in the range of blue light field is different from that in the yellow light field. It is the transmittance difference that caused the change of intensify ratio of blue light to yellow light in the SPD and leading to color change in perception. This indicates that the second optical material itself, even non-aged, could theoretically impact the color of the luminaire.

Figure 6b) shows the transmittance spectra of PMMA under different aging conditions. The values are almost the same and remain ~92%, which explained the phenomenon observed in Figure5 that relative luminous intensity slightly decreased both in the wavelength of blue light and in the yellow light area after PMMA (aged or not) was mounted and no significant color shift under different aging conditions, 85deg.C, additional exposure to blue light irradiation or humidity of 85%RH.

2.3.3 FTIR analysis

It is the inconsistent degradation of wavelength- dependent transmittance that gives rise to the SPD change and leads to the color shift of the luminaire using BPA-PC as the secondary optical design material. Hence, investigation on the cause of transmittance degradation will help to further understand the profound mechanisms of color shift. Accordingly FTIR studies were performed in order to find structural effects of aging stress on BPA-PC and PMMA materials respectively.

2.3.3.1 FTIR results of BPA- PC

Infrared Spectra of BPA-PC specimens with different aging time are shown in Figure7.

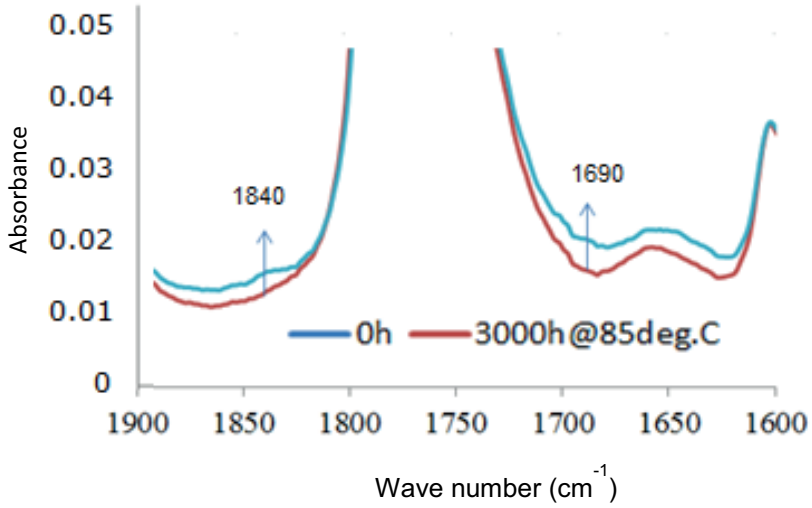


***Figure** 7: Infrared spectra of BPA-PC*

Results demonstrated that the band intensities increased at the wave number 1840cm^{-1} and 1690cm^{-1} which are associated with ketone and cyclic anhydride respectively, inferring that the oxidation had already taken place during thermal aging of 85deg.C up to 3000 hours, which is consistent with Yazdan Mehr's previous experiments [13-14] of thermal aging up to 3000 hours at 100~140deg.C for the same type of commercial BPA-PC. Side chain and ring oxidation as well as the formation and subsequent oxidation of phenolic end

groups are postulated to be the main reasons for discoloration and yellowing of thermally-aged BPA-PC [13-16]. Generally, oxidation plays a key role in the degradation of transmittance at around the peak wavelength of blue light field, which is in correlation with the discoloration of thermally-aged BPA-PC materials.

2.3.3.2 FTIR results of PMMA

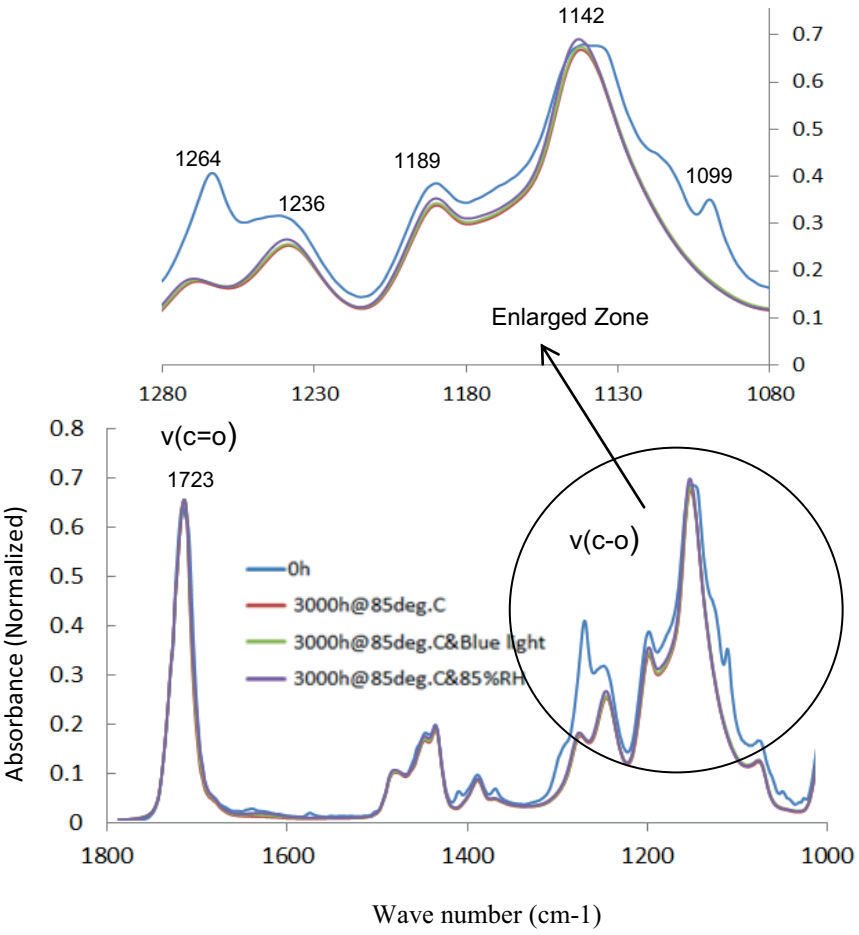


Figure 8: *Infrared spectra of PMMAs submitted to different aging* conditions

PMMA specimen submitted to different aging conditions were respectively

measured by FTIR, normalized results are shown in Figure 8. The C=O stretching vibration of ester group appears at around 1725 cm^{-1}, bands ranging from 1000 to 1300cm^{-1} correspond to the C-O stretching vibration [17-21].

There is no significant difference in the intensity of each individual band between aging conditions of 85deg.C, with additional exposure to blue light irradiation and additional humidity of 85%RH up to 3000 hours. That's probably the blue light exposure is not sufficient and the humidity has little impact on the degradation of the PMMA materials.

As mentioned previously, there is a pre-existing surface protective tape attached to the surface of the raw PMMA material. Before submitted to the aging stress, the protective tape was removed but the epoxy agent, which is rich in C-O bond, still existed on the surface of PMMA specimen. Such epoxy agent can be easily volatilized after 3000hours of aging. By contrast, there is still resident epoxy agent on the surface of PMMA for non-aged specimen during the FTIR measurement. The IR intensities of non-aged specimen at bands of 1264 and 1236cm^{-1} are much higher than those of specimen submitted to aging stress. Much likely, the additional FTIR intensity attributes to the resident epoxy agent on the surface of non-aged PMMA specimen. That is to say, there is probably no difference between the non-aged specimen and the aged ones excluding the epoxy agent.

Furthermore, the FTIR spectra of the specimen submitted to aging stress in the range of 1000~1800cm-1investigated in the paper is exactly the same as that of the virgin PMMA reported by Namouchi [17]. Hence, for the PMMA specimen investigated in this paper, it could be drawn that oxidation was neither occurred at 85deg.C, nor with additional exposure to blue light, nor even with additional humidity of 85%RH up to 3000hours. However, Estupinan et al reported the transmission change (1.01% decrease at 450nm) even after 90hours of aging with 72deg.C [22], different from results in this paper. Possible reasons are (1) PMMA type difference and (2) thickness impact. Estupinan's PMMA type is commercial PLEXIGLAS® df23 8N, while PMMA sample used in this paper is pure according to FTIR analysis; our sample is 3 mm thick, not yet find the thickness in Estupinan's publication. As our next step, thermal aging temperature will be increased to 100deg.C close to the Tg (around 110deg.C) of this material and different thickness and type of PMMA samples will be

included in order to further investigate the color shift effects and mechanisms with the same methodology used in this paper.

2.4 Conclusions

Broadly used BPA-PC and PMMA materials in the secondary optical designs combined with an LED package are experimentally investigated on the color shift effects of aging stress and color shift mechanism of degradation of transmittance. The conclusions could be drawn as follows:

1) Submitted to thermal aging of 85deg.C up to 3000hours, the discoloration and yellowing was observed for BPA-PC materials, while it didn't appear for PMMA materials under any implemented aging stress up to 3000 hours including 85deg.C, with additional exposure of blue light or additional humidity of 85%RH.

2) Inconsistent degradation of wavelength–dependent transmittance caused by the nature of oxidation process induces the decrease of the blue/yellow light intensity ratio and thus leading to the color shift toward the yellow field, which is the color shift mechanism of BPA-PC.

3) Even for the non-aged BPA-PC, the transmittance varies with wavelength in the visible light field due to the chemistry of the material，which caused the change of intensify ratio of blue light to yellow light in the SPD, leading to color change in perception. This indicates that the second optical material itself, even non-aged, could fundamentally impact the color of the luminaire.

4) Oxidation plays a key role in the degradation of transmittance at around the peak wavelength of the blue light field, which is in correlation with the discoloration/yellowing of thermally-aged BPA-PC materials. By contrast, for the PMMA specimen aged up to 3000 hours, oxidation was neither occurred at 85deg.C, nor with additional exposure to blue light, nor even with additional humidity of 85%RH.

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Chapter 3

Color Shift and Mechanism Investigation on the PMMA Diffuser Used in LED-based Luminaires[2]

PMMA diffuser material is widely used in LED-based luminaires due to several advantages such as excellent optical transparency, durability against radiation, surface hardness (scratch free), rigidity and strength and can be completely recycled. However, few studies have been reported on the color shift and failure mechanisms caused by this type of material. This paper experimentally investigated PMMA materials with different aging conditions. The following conclusions could be drawn: 1) Discoloration was not observed for any sample subjected to aging of 85℃ for 5000 hours, or with additional blue light irradiation for 5000 hours, or with additional humidity of 85%RH for 5000 hours, or even with aging of 100℃ for 3000 hours. 2) The specimen subjected to aging of 150℃for 360 hours has a surface discoloration and has a significant wavelength dependent degradation in the transmission spectrum caused by oxidation. The specimen with aging of 100℃ for 3000 hours has a less oxidation, although no significant transmission spectrum reduction was observed. 3) Using such aged specimen as a diffuser mounted on a LED-based luminaire, the radiant flux peak intensity in the blue light area has a more severe reduction than that in the yellow light area, which induces the color shift to yellow.

[2] This chapter is derived from the publication: Guangjun Lu, W.D. van Driel, Xuejun Fan, M. Yazdan Mehr, Jiajie Fan, Cheng Qian, K.M.B. Jansen, G.Q. Zhang. Color shift and mechanism investigation on the PMMA diffuser used in LED based luminaires. Opt. Mater. 54 (2016) 282–287

3.1 Introduction

Solid state lighting products are on the way of replacing the traditional incandescent lightings due to many advantages and continuous remarkable progress made recently on lighting efficacy, cost reduction, and quality improvement in mass production. Luminous flux maintenance and color stability are two important factors to evaluate the long term reliability, which are always concerns of some potential consumers and hence prevent use in some degree [1-4]. ENERGY STAR requires that the change of chromaticity over the lifetime of the product shall be within 0.007 on the CIE 1976 (u', v') diagram [5]. With increased adoption and accumulation of hours of use, awareness is growing that color shift is an issue which could not be neglected for solid state lighting especially in some important application fields, such as museums, patient examination rooms, urban scene illumination occasions, offices, street lighting and so on [6-7].

Color shift results from changes in spectral power distribution (SPD), including amplitude, peak wavelength and the shape of spectrum. Unlike traditional incandescent lighting products, color shift mechanisms of LED lighting are complex due to much more comprehensive structure, generally composed of LED dies, packages, lenses, reflectors, diffuser, electrical driver, and so on, and each of the individual parts may limit the long term stability and contribute to the color shift during operation [8].

In terms of color shift on the die or package level and even lenses, some studies have been done [9-12]. Yazdan Mehr et. al. experimentally investigated the color shift of remote phosphor plates made from Bisphenol-A polycarbonate (BPA-PC). The remote-phosphor plates were thermally aged at a temperature ranging from 100 to 140 ºC and some samples were exposed to blue light generated by blue LED sources. Significant decay was observed both in the phosphor yellow emission and in the blue peak intensity. The results also show that the degradation of the remote phosphor plates affects the efficiency of light and the color of emitted light as well. The decrease of CCT takes place with almost the same kinetics as the lumen depreciation [13-14]. Further chemical analysis illustrated that BPA-PC samples have turned into yellow in terms of

color appearance, inferring that the surface of the BPA-PC has been oxidized [15]. Similarly, M. Meneghini et al presented extensive analysis of the degradation of remote phosphor plates for application in LED-based light sources. Results indicate that when phosphor plates are submitted to irradiation with blue light, they can reach temperatures in excess of 60 °C and high temperature stress can result in a strong decrease in the efficiency of the phosphors, and in the change in the correlated color temperature [16]. Yanagisawa et al performed a current accelerated test on a commercial white LED at the temperature of 40℃ with humidity of 90%RH. It was observed that both peaks of SPD (the 460-nm emission peaks from the LED element and the 550-nm peaks from the excited fluorescent material) gradually decreased and the shift next to the long wavelength of the peak was 2nm after $1\sim2\times10^3$ h [17]. Trevisanello et al did thermal aging experiments on the 1W high-power LEDs with flip-chip configuration attached to a copper frame that operates as heat sink. The plastic white package is located around the chip and acts like a reflector cup. YAG phosphors for yellow conversion are situated inside the cup. The thermal aging induced a modification in the spectral shape in terms of yellow efficiency. Absolute measurements detected the lowering of both peaks and the degradation of yellow emission was more enhanced than the blue one. It was inferred that the change of spectral shape was potentially caused by 1) the lowering of phosphor efficiency; 2) the browning of the lens; and 3) the degradation of the package [18]. Wang et al presented the mean-time-to-failure (MTTF) evaluation of encapsulation materials of LED package in accelerated thermal tests. The results showed that the glass as encapsulation material of LED modules exhibited better thermal stability than the silicone encapsulation by 2 times in chromaticity shift [19].

However, literature on the color shift mechanism of LED-based luminaire level is little publicly available. Normally, reflectors and diffusers are mounted in the luminaire as secondary optical components, which are usually used to optimize the light extraction process and improve the light efficiency. However, they are susceptible to thermal stress and blue light irradiation during operation. Hence it is necessary to investigate the failure mechanisms and color shift effects induced by such secondary optics, which will help understand the whole luminaire color shift mechanisms and effects. According to Lu's investigation on the MCPET based reflector, the color shift is less than 0.001 even when the

side wall reflective material is scratched [20]. Color shift induced by the diffuser depends on the degradation of the transmission spectrum. Historical results show that the BPA-PC has much impact on the color shift due to oxidation. PMMA material is also widely used to make such a diffuser plate since it has several advantages such as excellent optical transparency, durability against radiation, surface hardness (scratch free), rigidity and strength and can be completely recycled [21]. The Molecular structure of PMMA is shown in Figure 1. Although it is reported in the reference [22] that the transmittance of PMMA specimens investigated was not changed after being subjected to 85℃and even with additional blue light and humidity for 3000hours, still some concerns exist since the claimed life for most LED-based luminaire suppliers is as long as over 20,000 hours, during which no one knows what the exact failure mechanisms could be and how much the color shift will be induced. This paper will report the results of more extremely accelerated experimental conditions including the failure mechanism and color shift effects which will be helpful to the design of secondary optics for LED-based luminaires.

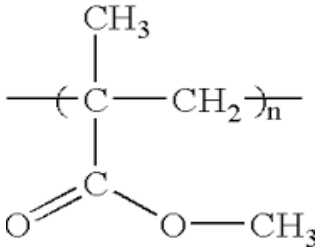


Figure 1 *Molecular structure of PMMA*

3.2 Materials and Methods

Commercial pure PMMA plates of 3mm thickness and with a dimension of 30mm×50mm, were submitted to the following experimental conditions respectively: 1) thermal aging in the oven with a temperature setting of 85℃ for 5000 hours 2) blue light irradiation at 85 ℃ for 5000 hours (The specimens were placed inside the oven with a setting of 85℃ and exposed to the blue light through the oven glass window generated by the blue light LEDs outside the oven. The blue light intensity that the specimen received is around 40k lux, and

the peak wavelength of the blue light source is 450nm. (Refer to [22] for more details) 3) humidity test for 5000 hours with a temperature setting of 85℃ and relative humility of 85% RH. In addition, specimens of the same type and thickness with a dimension of 100mm×100mm were put separately in two ovens with a temperature of 100 ℃ for 3000 hours and 150℃ for 360 hours respectively. Transmittance spectra of the specimens under different aging conditions were recorded in the range 380~740 nm with a step of 5nm with the Lambda 950 spectrophotometer (PerkinElmer 950). Infrared spectra of specimens were measured for chemical analysis in the range of 500–4000 cm^{-1} using a Perkin–Elmer Spectrum 100 series spectrometer in the attenuated total reflection (ATR) mode for 700 scans at a resolution of 0.5 cm^{-1}.

In order to quantify the color shift effects of PMMA specimens after aging, a down-light LED luminaire was used and measured by an integrating sphere with a diameter of 2m at 2pi mode. Measurement errors have been minimized by a strict control on the major error contributors as follows

1) The measurements were always performed by a same person;
2) Each time in the measurement, the luminaire was mounted and aligned to the same position;
3) The measurement values shown in this paper are the averages of 3 measurements.



Figure 2 *PMMA specimen mounted on the luminaire for integrating sphere measurement*

Prior to measurement, the PMMA specimens were mounted onto the luminaire, as shown in Figure 2. LED packages attached in the luminaire are mid-power 5630 with a CCT of 4000 k, whose spectral power distribution (SPD) is shown in Figure 3, which is from the LED package datasheet [23].

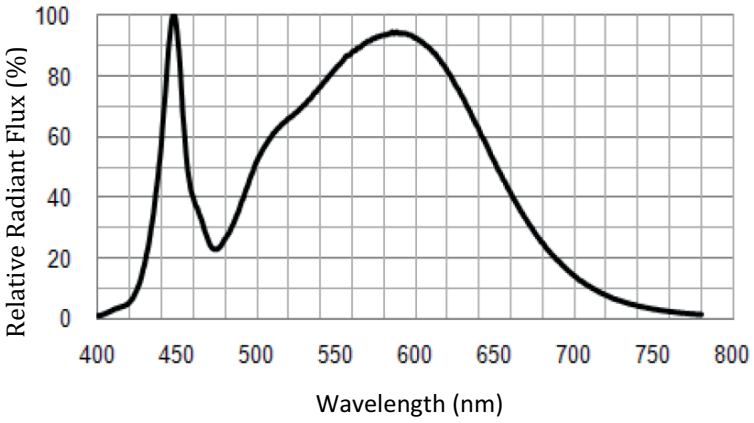


Figure 3 *SPD of LED packages used in the LED-based luminaire*

3.3 Results and Discussion



a) Non-aged b) Aged with 150℃ for 360h

Figure 4 *Discoloration of PMMA after aging*

Discoloration was not observed for any sample subjected to aging of 85℃ for 5000 hours, or with additional blue light irradiation for 5000 hours, or with additional humidity of 85%RH for 5000 hours, or even with aging of 100℃ for 3000 hours. By contrast, after 360 hours of thermal aging at 150℃ in the oven, the color in the surface of PMMA specimen tends to be slightly yellowing. Pictures of the specimen before and after aging are shown in Figure 4. Note that the aging temperature 150℃ is above Tg (around 110℃) and below melting point (around 200℃) of the PMMA material, and we didn't observe any color change of the surface during the first day of aging.

3.3.1 Transmission spectra measurements

The transmission spectra of specimens subjected to different aging conditions were measured and recorded respectively by the spectrophotometer Lambda 950 in a wavelength range from 360nm to 760nm with a step of 5nm. Results are shown in Figure 5. The transmittance of specimen aged at 150℃ for 360 hours is lower than that of any subjected to other aging conditions in the wavelength range from 360 nm to 760 nm, and the specimens subjected to other aging conditions have almost the same spectra as that of the non-aged specimen.

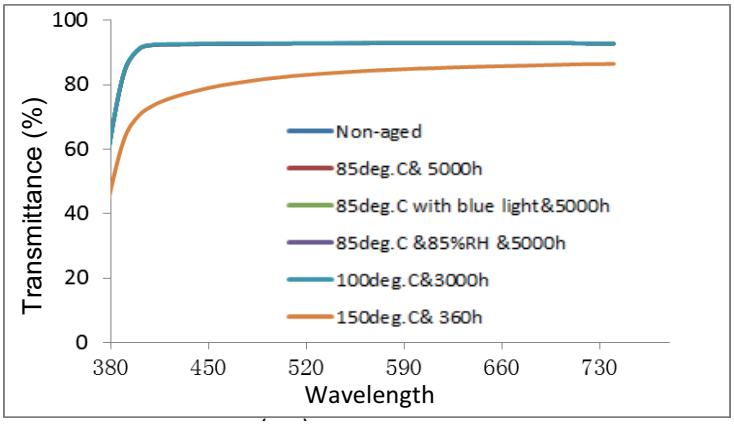


Figure 5 *Transmission spectra after being subjected to different aging conditions (Note: the 85℃ and 100℃ lines coincide with the non-aged line.)*

Furthermore, as shown in Figure 6, the transmittance reduction (calculated on the difference of transmittance between lines of non-aged and aged at 150℃ shown in Figure 5) of the specimen aged at 150℃ is wavelength dependent and

the reduction of transmittance at around 450nm (blue light area) is much higher than that at around 590nm (yellow light area).

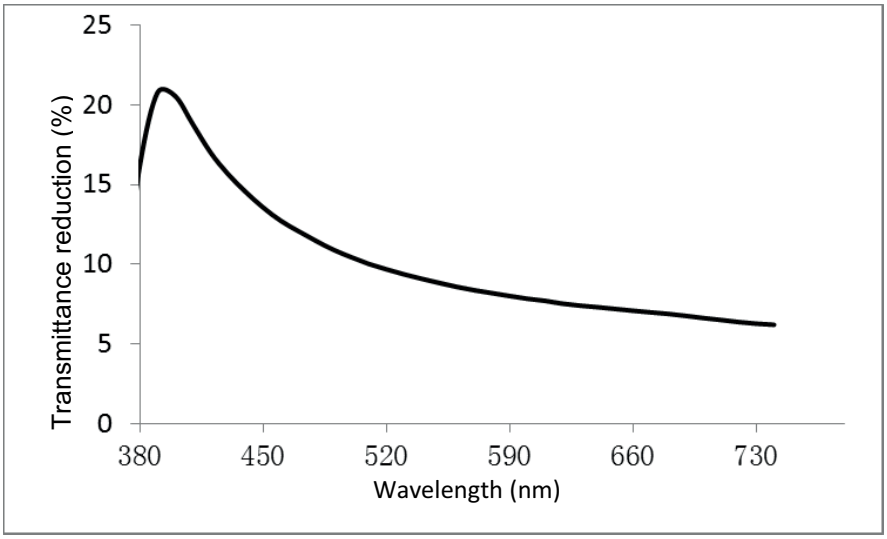


Figure 6 *Transmittance reduction for the PMMA subjected to 150℃ for 360h*

3.3.2 FTIR Analysis

FTIR studies were performed in order to chemically investigate the above mentioned transmission spectrum change after aging. Baseline correction was done during operation. The absorption spectroscopies, normalized on the peak of C=O stretching vibration (1720cm^{-1}) since the bond C=O is stable, are shown in Figure 7.

Bands ranging from 1000 to 1300cm^{-1} correspond to the C-O stretching vibration [24-27]. The specimen subjected to 150℃ aging has a significant increase in the peak intensities both at (1142, 1189cm^{-1}) and at (1240, 1269cm^{-1}), which implies oxidation occurred to that specimen. There are three steps involved in the aging. The first step is the splitting off of the acrylate group, whereas the second step is the actual oxidation and followed by hydrogenation [28]. Such chemical mechanism can be described in Figure 8.

In addition, absorption peaks are also observed in the same specimen with aging of 150℃ in the band ranging from 1500cm^{-1} to 1600cm^{-1}, which are associated with the asymmetric stretch vibration of the carboxyl –COOH

generated during oxidation. A slight peak increase at 1142 cm^{-1} is observed together with weaker peaks occurring in the range from1500cm^{-1} to 1600cm^{-1} implying that the specimen aged at 100℃ for 3000 hours has a less oxidation, although significant transmission spectrum changes were not observed. However, Estupinan et al reported the transmission change (1.01% decrease at 450nm) even after 90 hours of aging with 72deg.C [29], quite different from results in this paper. Probably the commercial PMMA type and thickness are different. PMMA sample used in this paper is pure according to FTIR results.

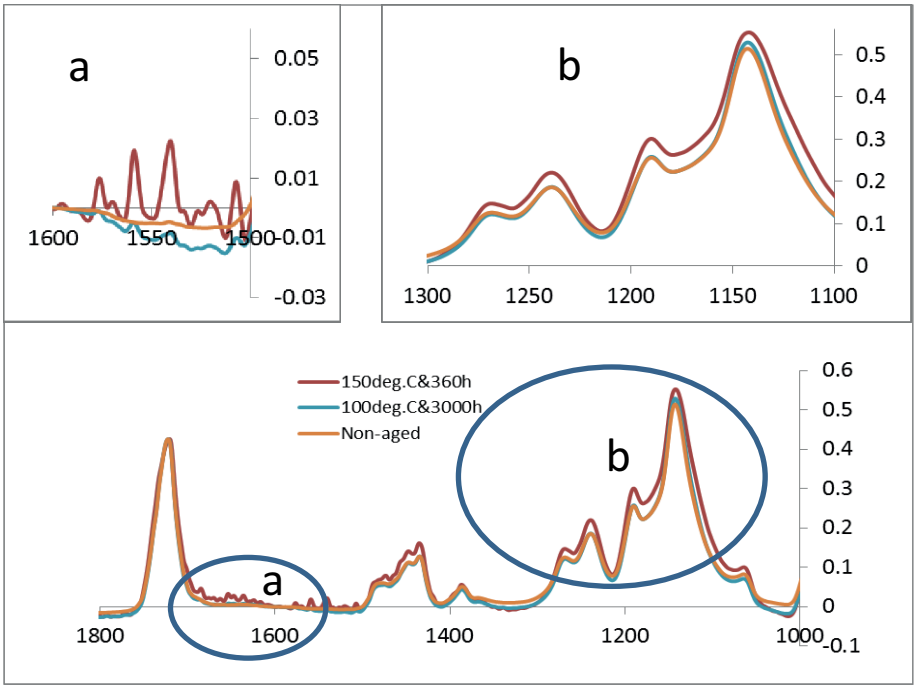


Figure 7 *Infrared absorption spectra of PMMA with different aging conditions:150 ℃ for 360h and 100 ℃ for 3000h*

It should be pointed out here the experiments found that PMMA will exhibit yellowing only at high temperatures (150℃) which are well above the glass transition temperature of the polymer. This temperature is also above the accepted use temperature of PMMA and is unlikely to be used in a practical luminaire. Therefore, the risks on introducing a new failure mode is high since the experiments were performed above Tg and chain mobility will be increased.

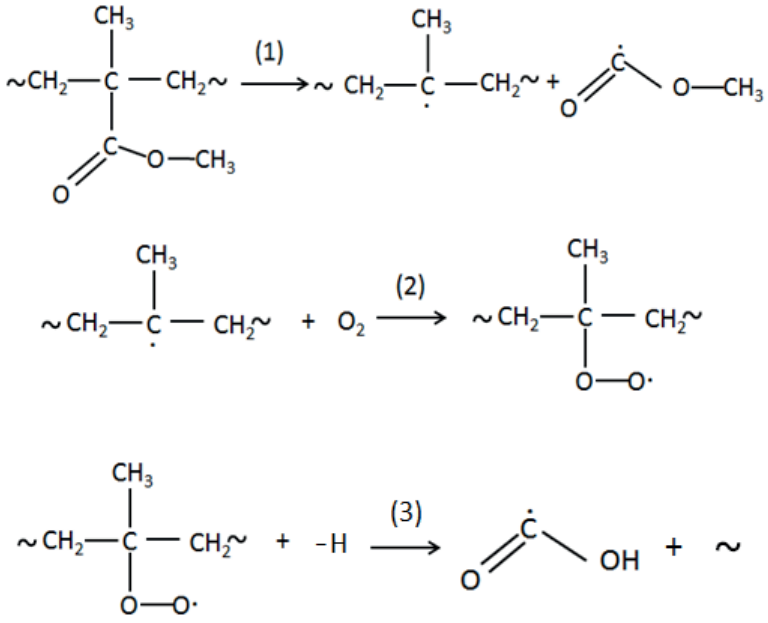


Figure 8 *Chemical mechanisms of oxidation and hydrogenation during aging*

3.3.3 Color shift effects investigation

The non-uniform reduction of transmittance in the transmission spectrum caused by aging could induce the change of radiant flux intensity ratio of blue light to yellow light, which gives rise to the color shift in perception and chromaticity change in the CIE1976 diagram. In order to quantify color shift effects of the specimen subjected to 150℃ for 360 hours, as aforementioned, a down-light LED luminaire mounted with such an aged sample and a non-aged one of identical size was measured by an integrating sphere. Normalized spectral power distributions are shown in Figure 9. Some fraction of the emission from the LED does not pass through the lens and undergo absorption as shown in the Figure2, which will contribute a little error to the measurement. The results of color coordinates in the CIE1976 diagram by such measurement are also given, as shown in Figure 10. As can be found in Figure 9 the peak wavelength neither

at blue light area (around 450nm) nor at yellow light area (around 590nm) changed; however, the peak intensity at blue light area has a more severe reduction than that at the yellow light area, which causes the reduction of radiant flux intensity ratio of blue light to yellow light and hence induces the color shift to yellow as confirmed in Figure 10. Color shift data is shown in Table 1.

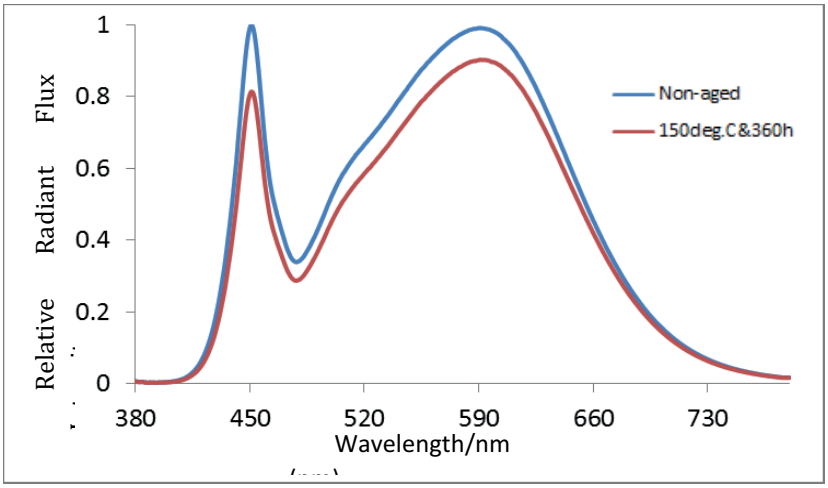


Figure 9 *SPDs of LED-based luminaire mounted with aged PMMA specimen*

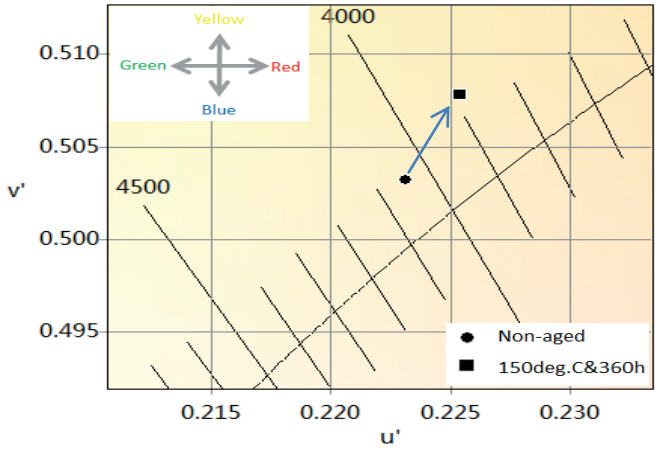


Figure 10 *Color coordinates in the CIE 1976 Diagram for the LED-based Luminaire mounted with PMMA specimens*

Table 1 Color shifts after being subjected to 150℃ for 360h

Aging Condition	CCT	u'	v'	Delta (u')	Delta (v')	Delta (u' v')
Non-aged	*4048*	*0.2231*	*0.5032*	/	/	/
150℃@360h	*3887*	*0.2254*	*0.5078*	*0.0023*	*0.0046*	*0.0051*

The CCT, results for u' and v' as shown in Table 1 were obtained directly from the integrating sphere after the measurement. The color data of the luminaire with non-aged PMMA specimen is taken as the reference (u_0', v_0'). The color shift Delta (u'), Delta (v') and Delta (u' v') shown in Table 1 are defined as follows:

$$\text{Delta}(u') = u' - u_0' \quad (1)$$

$$\text{Delta}(v') = v' - v_0' \quad (2)$$

$$\text{Delta}(u'v') = \sqrt{\left[\text{Delta}(u')\right]^2 + \left[\text{Delta}(v')\right]^2} \quad (3)$$

In addition, the luminous flux Φ(lm) data was also measured and shown in Table 2. The luminous flux of the luminaire mounted with a non-aged sample is defined as Φ_0(lm). The percentage of lumen decay Delta [Φ(lm)]% is calculated on the equation (4).

$$\text{Delta}[\Phi(\text{lm})]\% = [\Phi_0(\text{lm}) - \Phi(\text{lm})] * 100\% / \Phi_0(\text{lm}) \quad (4)$$

Table 2 Luminous flux decay after being subjected to 150℃ for 360h

Aging Condition	Power P(W)	Lumen Φ(lm)	Lumen decay Delta[Φ(lm)]%
Non-aged	*10.12*	922.64	/
150℃@360h	*10.12*	*828.36*	*10.22*

It can be found from Table 1 that the color shift of the luminaire induced by the aged PMMA diffuser is 0.005, very close to the general failure criterion of 0.007, although the lumen decay is only 10.2% as shown in Table 2, far less than the failure criterion of 30%, which implies that the PMMA color shift failure caused by thermal oxidation could probably occur prior to the failure due to lumen degradation.

3.4 Conclusions

The color shift and failure mechanism were investigated on the PMMA diffuser used in LED-based luminaire products. Conclusions could be drawn as follows:

1) No yellowing was observed for any sample subjected to aging of 85℃ for 5000 hours, or with additional blue light irradiation for 5000 hours, or with additional humidity of 85%RH for 5000 hours, or even with aging of 100℃ for 3000 hours. However, the surface discoloration occurred to the sample aged at 150℃ for 360 hours.

2) The specimen subjected to aging of 150℃ for 360 hours has a significant wavelength dependent degradation in the transmission spectrum, which is caused by oxidation according to the result of FTIR analysis. The transmittance reduction in the blue light area is higher than that in the yellow light area. FTIR analysis implies that the specimen with aging of 100℃ for 3000 hours has a less oxidation, although no significant transmission spectrum reduction was observed.

3) Using the PMMA specimen aged at 150℃ for 360 hours as a diffuser mounted on the luminaire, the radiant flux peak intensity in the blue light area has a more severe reduction than that in the yellow light area, which results in the reduction of radiant flux intensity ratio of blue light to yellow light and hence induces the color shift to yellow. The color shift investigated is 0.005, very close to the general failure criterion of 0.007, while the lumen decay is 10.2%, far less than the lumen decay failure criterion of 30%.

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Chapter 4

Degradation of Microcellular PET Reflective Materials Used in LED-based Products[3]

Microcellular PET is an emerging reflective material used for solid state lighting. This paper experimentally investigated its degradation mechanisms and quantified both lumen decay and color shift effects of LED-based products with this material aged under different conditions. The results show that: 1） A humidity test at 85 ºC & 85%RH for 4000hours (or even shorter) can lead to hydrolytic degradation, which causes both the decrease of reflectance varying with wavelength and a severe embrittlement; 2) Oxidative degradation occurred at 85 ºC for 4000hours can cause a slight reflectance spectrum change, while the additional blue light exposure has little impact; 3) Color shift induced by thermal aging at 85 ºC for 4000hours is 0.0001 and lumen efficiency decreased by 1.57%. When Microcellular PET crumbles due to the embrittlement during humidity test, the color shift increases to 0.0004 and the lumen efficiency is reduced by 4.47%.

[3] This chapter is derived from the publication: Guangjun Lu, W.D. van Driel, Xuejun Fan, M. Yazdan Mehr, Jiajie Fan, K.M.B. Jansen, G.Q. Zhang Degradation of Microcellular PET Reflective Materials Used in LED-based Products, Opt. Mater. 49 (2015) 79–84

4.1 Introduction

Solid state lighting products are constantly increasing their market share in the worldwide general lightings due to continuous remarkable progress made on lighting efficacy, cost reduction, and quality improvement and thus winning recognition from consumers. Lumen decay and color shift are two key failure modes in the application of LED-based products [1-2], which are always concerns of some potential consumers and hence limits the acceptance in some degree. A vast of studies have been done on the mechanisms of Lumen decay, however, few researches have been conducted on the color shift [3-5]. ENERGY STAR requires the change of chromaticity over the lifetime of the product shall be within 0.007 on the CIE 1976 (u',v') diagram [6]. With increased adoption and accumulation of hours of use, awareness is growing that color shift is an issue which could not be neglected for solid state lighting especially in some important application fields, such as museums, patient examination rooms, urban scene illumination occasions, offices, street lighting and so on [7-8].

Color shift results from changes in spectral power distribution (SPD), including amplitude, peak wavelength and the shape of spectrum. Unlike traditional incandescent lighting products, color shift mechanism of LED lighting is complex due to much more comprehensive structure, generally composed of LED dies, phosphors, packages, lenses, reflectors, diffuser, electrical driver, and so on, and each of individual part may limit the long term stability and contribute to the color shift during operation [9].

Some studies of color shift on the die and package level including lenses have been done [10-11]. M. Yazdan Mehr et. al. experimentally investigated the color shift of remote phosphor plates made from Bisphenol-A polycarbonate (BPA-PC). The remote-phosphor and lens of BPA-PC specimens of 3 mm thickness were thermally aged at temperature ranging from 100 to 140 ºC and some samples were exposed to blue light generated by blue LED sources. It was observed there is a significant decay both in the phosphor yellow emission and in the blue peak intensity. The decrease in the luminous flux is strongly correlated to the worsening of the chromatic properties of the phosphor plates. The results also show a significant decay of CCT, claiming that the degradation

of the remote phosphor plates affects the efficiency of light and the color of emitted light as well. The decrease of CCT takes place with almost the same kinetics as the lumen depreciation [12-13]. Further chemical analysis was done on the aged samples, illustrating that BPA-PC samples have turned into yellow in terms of color appearance, inferred that the surface of the BPA-PC has been oxidized [14].

T.Yanagisawa et al performed a current accelerated test on a commercial white LED with a structure based on an epoxy lens/YAG fluorescent substance/InGaN LED chip at the temperature of 40ºC with relative humidity of 90%RH. It was observed that both peaks of SPD (460-nm emission peaks from an LED element and the 550-nm emission peaks from excited fluorescent material) gradually decreased and the shift next to the long wavelength of the peak was 2nm after 1~2 ×103 h [15]. L. Trevisanello et. al. did thermal aging experiments on the 1-W HB LEDs with the structure of 1mm2 areas InGaN/GaN LED in flip-chip configuration attached to a copper frame that operates as heat sink. The plastic white package is located around the chip and acts like a reflector cup. YAG phosphors for yellow conversion are situated inside the cup. The thermal aging induced a modification in the spectral shape in terms of yellow efficiency. Absolute measurements detected the lowering of both peaks and the degradation of yellow emission was more enhanced than the blue one. It was inferred that the change of spectral shape was potentially caused by 1) the lowering of phosphor efficiency; 2) the browning of the lens; and 3) the degradation of the package [16]. Also, different types of phosphor have different impacts on chromaticity shift [17-18]. J. Wang et. al. presented the mean-time-to-failure (MTTF) evaluation of encapsulation materials of LED package in accelerated thermal tests. The results showed that the glass as encapsulation material of LED modules exhibited better thermal stability than the silicone encapsulation by 2 times in chromaticity shift [19]. M. Meneghini et al presented extensive analysis of the degradation of remote phosphor plates for application in LED based light sources. Results indicate that when phosphor plates are submitted to irradiation with blue light, they can reach temperatures in excess of 60°C and high temperature stress can result in a strong decrease in the efficiency of the phosphors, and in the change in the correlated color temperature [20].

However, literature on the color shift mechanisms of LED based luminaire level is little publicly available. The luminaire reflector is an important component embedded in the luminaire, which is usually used to optimize the light extraction process and impact the light efficiency and is susceptible to the thermal stress and blue light irradiation during operation. Hence it is necessary to investigate the failure mechanisms and color shift effects induced by the embedded reflector, which will help understand the luminaire color shift mechanisms and effects. Microcellular PET (Polyethylene terephthalate) is being widely used as the reflective material for LED–based luminaires. Compared to the traditional metal-based reflective material, the Microcellular PET has many advantages, such as high total reflectivity, high diffuse reflectivity，and light weight [21]. However, previous research on the failure mechanisms and color shift effects of Microcelluar PET applied as the LED reflective material is quite few. Thus, the motivation of this paper is to experimentally investigate the failure mechanisms and quantify color shift effects under different aging conditions based on Microcellular PET material for solid state lighting, which will pave the way for the development of luminaire level color shift acceleration method in the future. The Molecular structure of PET is shown in Figure 1. Figure 2 shows the schematic diagram and principle of Microcellular PET reflective materials. SEM images of cross section of a Microcellular PET specimen are shown in Figure 3.

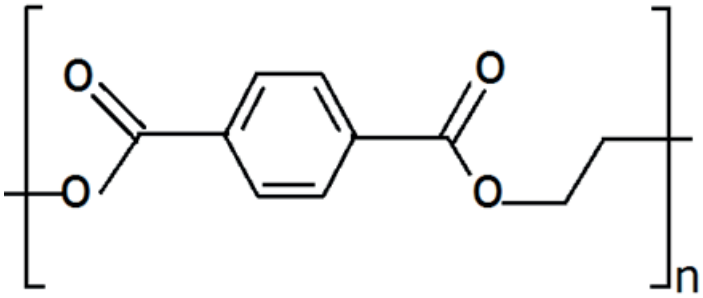


Figure 1 *Molecular structure of Polyethylene terephthalate*

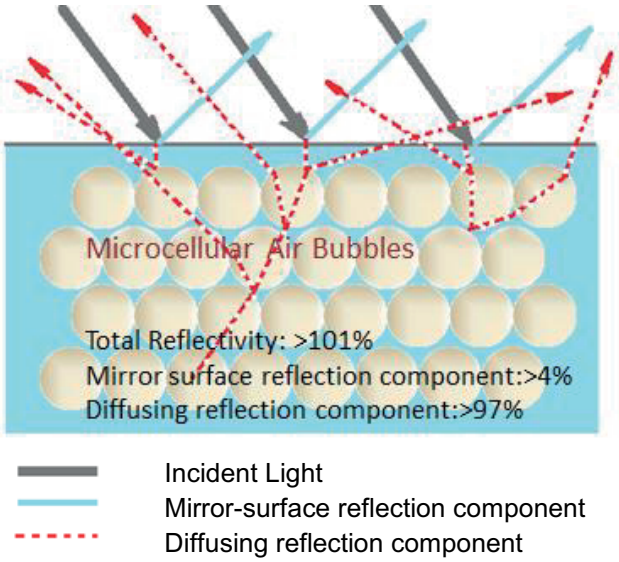


Figure 2 *Schematic diagram and principle of Microcellular PET reflective materials*

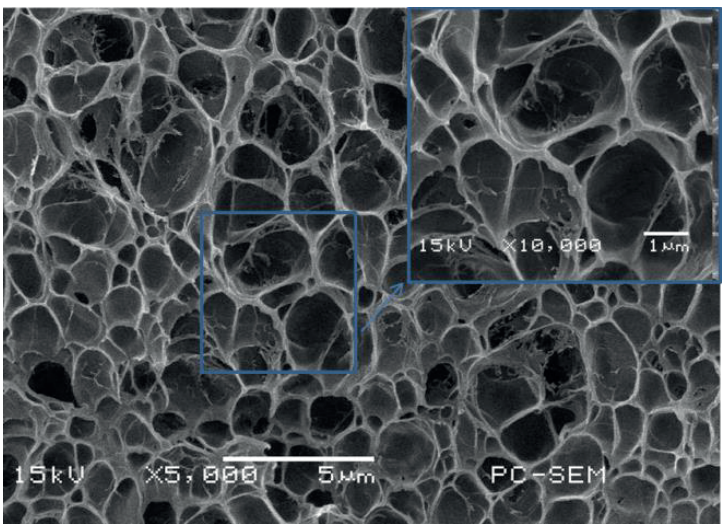


Figure 3 *SEM images of cross section of a Microcellular PET*

4.2 Materials and Methods

Thickness of 0.51mm of commercial rectangular Microcellular PET reflective sheets with a dimension of 30mm×50mm, were subjected to the following 3 experimental conditions respectively:

1) Thermal aging in an isothermal oven with a temperature setting of 85 ºC for 4000 hours;

2) Blue light irradiation at 85 ºC for 4000hours (The specimens were placed inside the oven at 85 ºC and exposed to blue light through the oven glass window generated by the blue light LEDs outside the oven. The wavelength of blue light used is 450nm, the distance between blue light source and the specimens is 40cm and the blue light intensity that the specimen received is around 40k lux. Refer to [22] for more details);

3) Humidity reliability test (Specimens were put in the isothermal oven for 4000 hours at 85 ºC and 85% RH).

Reflectance spectra of specimens under different aging conditions were recorded in the range 380~800 nm with a step of 5nm with the Lambda 950 spectrophotometer (PerkinElmer 950). The UV/vis wavelength range calibration was done before measurement. During the calibration, the routine of default setting of auto search 2 peaks was chosen. The calibration software performs an automatic search for the D2 peak at 656.1nm and the peak at 0.0nm. The measured peaks are then shifted to the exact wavelengths. Relative reflectance mode other than the absolute mode is chosen to do the measurement with a universal reflectance accessory as the light spectral reference. All the reflectance measurements reported in this paper are relative to the reflectance of the UV/vis spectrometer's integrating sphere.

Stress-strain curves were also measured with a DMA Q800 to characterize the mechanical properties of specimens with different aging conditions. Infrared spectra of specimens were measured for chemical analysis in the range of 900–1800 cm^{-1} using a Perkin–Elmer Spectrum 100 series spectrometer in the attenuated total reflection (ATR) mode for 180 scans at a resolution of 5 cm^{-1}.

In addition, in order to quantify the color shift effects of Microcellular PET specimens after aging, a down-light LED luminaire was used and measured by

an integrating sphere with a diameter of 2m under 2pi mode. Measurement errors have been minimized by a strict control on the major error contributors as follows

1） Only one person did the measurement, no someone else was involved in the measurement;
2） Every time in the measurement, the luminaire was mounted and aligned to the same direction;
3） The measurement values shown in this paper are average values of 3 times of measurement.

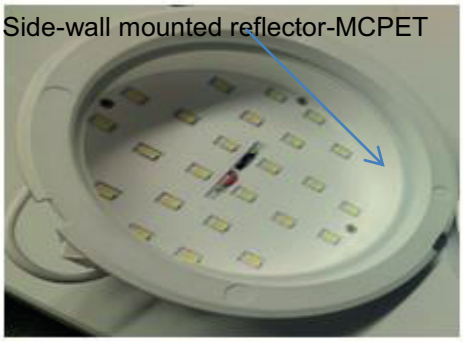


Figure 4 Down- light LED luminaire used for color shift Investigation

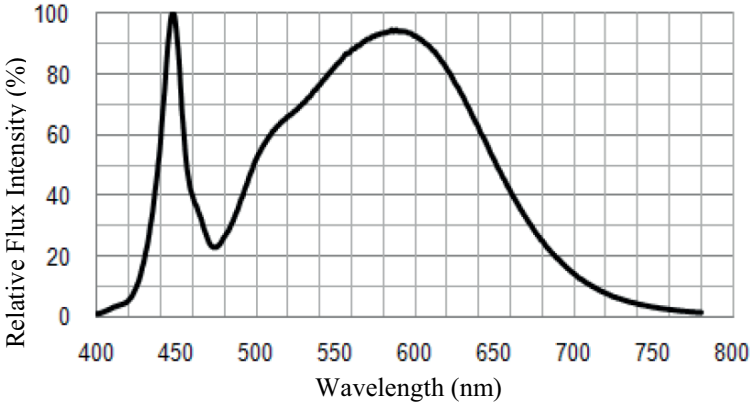


Figure 5 *SPD of LED packages used in the down-light LED luminaire*

Before the measurement, the Microcellular PET material specimens were cut and mounted into the luminaire as side-wall reflectors respectively, as shown in Figure 4. LED packages attached in the luminaire are mid-power 5630 with a CCT of 4000 k, whose spectral power distribution (SPD) is shown in Figure 5, which is from the LED package datasheet [23].

4.3 Results and Discussions

After 4000hours of aging, discoloration was not observed to any specimen. However, during manually handling when we took out the samples from the humidity test oven, one specimen broke as shown in Figure 6, which is probably because the mechanical strength of the sample become weakened after humidity test. Hence, the stress-strain measurement was conducted on all the specimens to investigate the possible changes of mechanical property in spite of reflectance spectrum measurements for investigation on the optical property changes.



Figure 6 *Specimen smashed after 4000 hours of humidity test during handling*

4.3.1 Reflectance spectra measurements

The reflectance spectra of specimens after 4000 hours of aging were measured and recorded by a Lambda 950 spectrophotometer in a wavelength

range from 380nm to 800nm with a step of 5nm. Results are shown in Figure 7. The reflectance spectrum of the 85℃ aged specimen is nearly the same as that exposed to additional blue light irradiation. Furthermore, the reflectance spectrum of the 85℃ aged specimen is higher than the non-aged one in the wavelength range from 380nm to 430 nm. The specimen which was subjected to humidity test has an obvious reflectivity decrease in the range from 380nm to 520nm compared to the non-aged sample. In addition, the reflectance spectrum of one type side-wall base material for luminaire was measured as shown in Figure 7, is lower than Microcellular PET for any type of aging or non-aging in the whole visible light wavelength range.

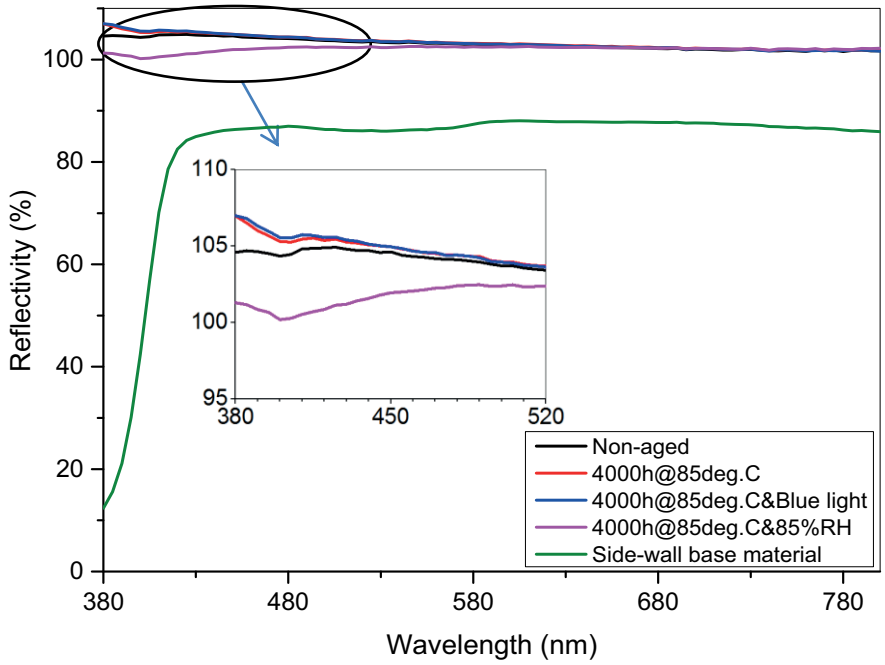


Figure 7 *Reflectance spectra of Microcellular PET with different aging conditions and side-wall base material*

4.3.2 Stress-strain measurements

In order to investigate the mechanical property changes of the aged specimens, rectangular strips with a dimension of 4mm x 30mm were cut from the specimens to do stress-strain measurement with a DMA Q800 machine. Samples which underwent 4000 hours of humidity were too brittle and fractured

during sample preparation. We had to use the 3000 hours of humidity test samples instead for the stress-strain tests. Typical stress-strain curves are shown in Figure 8. The samples were stretched at a speed of 0.1mm/min. Only the sample subjected to humidity test fractured and this fracture occurred at a strain of 0.38% (1.1Mpa). The samples of the non-aged and 4000 hours thermally aged did not fracture during the test. It can also be observed that the non-aged sample shows a clear ductile behavior with a yield stress of about 6Mpa.

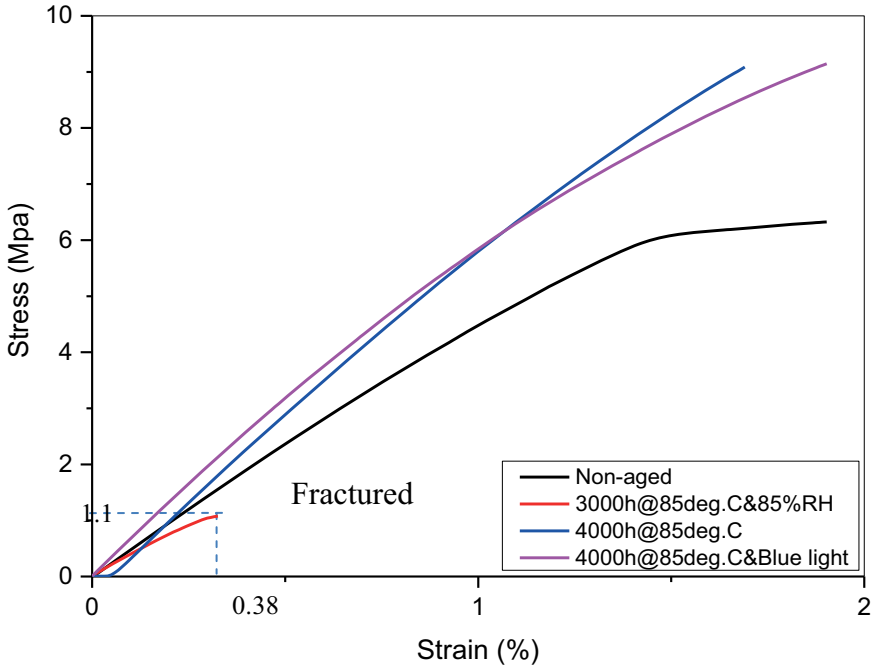


Figure 8 *Stress-strain relations of Microcellular PET with different aging conditions*

4.3.3 FTIR Analysis

FTIR studies were performed in order to chemically explain the above mentioned optical and mechanical property changes after aging. Baseline correction was done during operation. The absorption spectroscopies, normalized on the peak of C=O stretching vibration (1720cm^{-1}), are shown in Figure 9.

The specimen with humidity test has significant increase in the peak intensities of band 1340cm^{-1} and band 1410cm^{-1} which are associated with in-

plane bending vibration of -O-H in the carboxyl group. According to early investigations on the hydrolytic degradation of PET, such carboxyl group can be increased by hydrolytic ester scission [24]. The chemical reaction mechanism of hydrolytic degradation can be described in Figure 10 [25-26].

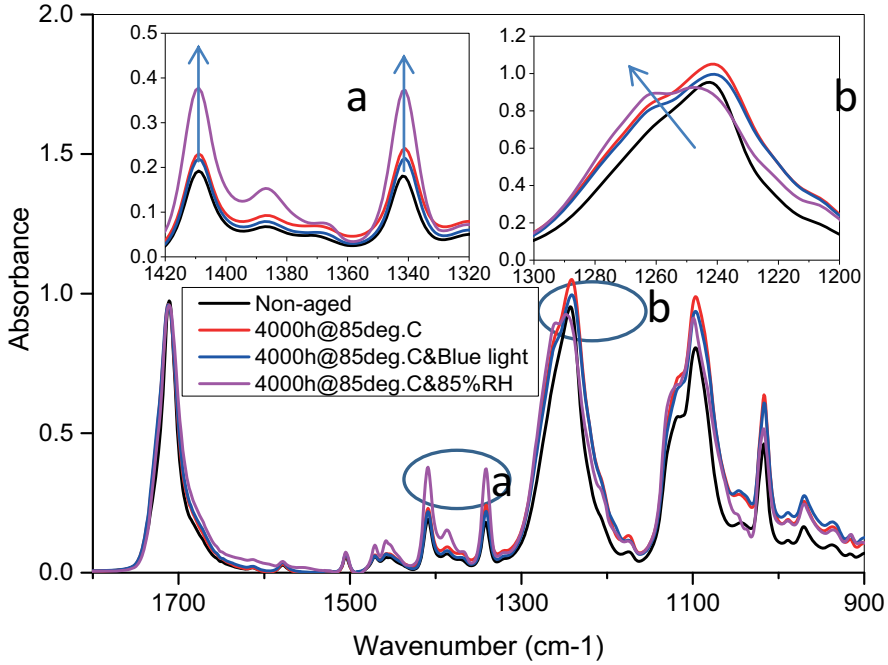


Figure 9 *Infrared absorption spectra of Microcellular PET with different aging conditions: Top left is the extended area of a, and top right is the extended area of b*

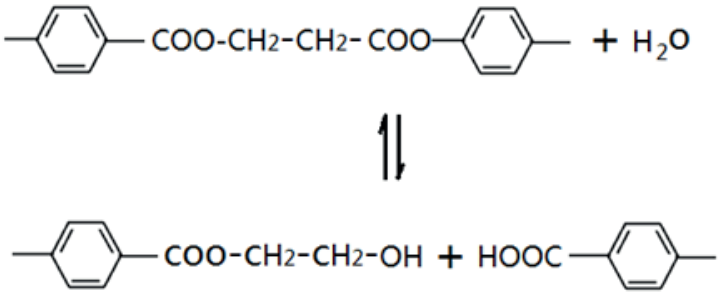


Figure 10 *Mechanism scheme of hydrolytic degradation of Microcellular PET*

Hydroxyl end-groups increased during hydrolytic degradation induces chemical shift at 1240cm^{-1} band (C-O stretching vibration) to the higher wave number 1262cm^{-1}, which can be observed in the top right for the extended area of b in Figure 9. Since such hydroxyl end-groups can be also generated during thermal oxidative degradation [22-23], specimen with thermal aging of 85 ºC has a similar but slight chemical shift as shown in Figure 9. This implies that the thermal oxidative degradation of PET is slower than the hydrolytic degradation at 85 ºC, which is in agreement with the early conclusion based on the degradation temperature of 100~125 ºC [25-26].

Comparing to the thermally aged sample, specimen with additional blue light irradiation did not show significant difference in the absorption spectroscopy， which implies that blue light did not have any extra contribution on aging of specimens, probably due to the insufficient exposure or PET's lack of sensitivity to blue light, similar result can be found in the PMMA degradation[22].

Hence, compared to the slower oxidation in the thermal aging test, the hydrolytic degradation in the humidity test induces chemical structure change of the specimen, which causes the remarkable changes of optical and mechanical properties.

4.3.4 Color shift and lumen decay investigation

The inconsistent degradation rate of reflectivity in the reflectance spectrum caused by aging could induce the change of luminous intensity ratio of blue light to yellow light, which gives rise to the color shift in perception and chromaticity change in the CIE1976 diagram. In order to quantify color shift effects of those specimens after aging under different conditions, as aforementioned, a down-light LED luminaire with aged and non-aged specimens was measured by an integrating sphere respectively. The embrittlement of the microcellular PET after 4000 hours of humidity test will make it so fragile that when it is disturbed (e.g., for repair or ceiling maintenance) the material may crumble exposing the underlying paint reflector. Consequently, to investigate the color shift effects due to such possible crumbling for 4000 hours (or longer) of humidity test, the Microcellular PET was not mounted, during which the side-wall base material, which has a lower reflectivity, act as a reflector. Color shifts are shown in Table 1.

The item CCT, u' and v' shown in Table 1 were got directly from the integrating sphere after measurement, and assumed that the color data of the luminaire with non-aged MCPET is the central color (u_0', v_0'), the color shift Delta (u'), Delta (v') and Delta (u' v') shown in Table 1 are derived from the equations as follows:

$$\text{Delta}(u') = u' - u_0' \quad (1)$$

$$\text{Delta}(v') = v' - v_0' \quad (2)$$

$$\text{Delta}(u'v') = \sqrt{\left[\text{Delta}(u')\right]^2 + \left[\text{Delta}(v')\right]^2} \quad (3)$$

Table 1 Color shifts after subjected to different aging conditions

Aging Condition	CCT	u'	v'	Delta (u')	Delta (v')	Delta (u' v')
on-aged	*4071*	*0.2229*	*0.5023*	/	/	/
4000h@85℃	*4073*	*0.2228*	*0.5023*	*0.0001*	*0*	*0.0001*
4000h@85℃&85%RH	*4085*	*0.2226*	*0.502*	*0.0003*	*0.0003*	***0.0004***

It can be found in Table 1 that the specimen subjected to 4000 hours of thermally aged test has a color shift of 0.0001 due to a reduction of u' by 0.0001, while 4000 hours of humidity test causes a color shift of 0.0004 with an equivalent reduction of u' and v' of 0.0003.

In addition, the lumen maintenance data was also measured and shown in Table 2. The Luminous Efficacy (LE), Delta (LE) and Lumen Efficiency Change (LEC) shown in Table 2 are calculated on the equations (4-6). LE_0 is assumed to be the Luminous Efficacy of non-aged specimen.

Table 2 Luminous maintenance data after subjected to different aging conditions

Aging Condition	Power P(W)	Lumen Φ(lm)	Luminous Efficacy LE(lm/W)	Delta(LE) (lm/W)	Lumen Efficiency Change LEC (%)
Non-aged	*10.11*	*956.56*	*94.62*	/	/
4000h@85℃	*10.11*	*941.50*	*93.13*	*-1.49*	*-1.57*

4000h@85℃ &85%RH	*10.11*	*913.81*	*90.39*	*4.23*	***-4.47***

$$LE = \Phi(lm) / P(W) \quad (4)$$

$$Delta(LE) = LE - LE_0 \quad (5)$$

$$LEC(\%) = (LE - LE_0) * 100 / LE_0 \quad (6)$$

It can be found in Table 2 the lumen efficiency has a reduction of 4.47% when the reflective material Microcellular PET crumbles under some situation due to 4000 hours of humidity test, compared to a reduction of 1.57% when the specimen was aged at 85℃ for 4000 hours. This implies that the lumen efficiency will be improved and increased by 4.47% when the non-aged Microcellular PET is applied as the side-wall reflector compared to the bare side-wall base material with a lower reflectivity in the down-light LED luminaire.

4.4 Conclusions

Color shift and lumen decay under different aging conditions were investigated on the Microcellular PET reflective material with a down-light LED luminaire. Conclusions can be drawn as follows

1) Hydrolytic degradation of Microcellular PET under humidity test of 85 ºC and 85%RH with 4000hours (or even shorter) can cause both the decrease of reflectance varying with wavelength and a severe weakening of mechanical strength (embrittlement).

2) Under the thermal aging test at 85 ºC with 4000hours, oxidative degradation causes a slight reflectance spectrum change in the wavelength range from 380nm to 430nm. Additional blue light exposure has little impact on degradation, which is probably due to the insufficient exposure or PET's lack of sensitivity to blue light.

3) Optical measurement results indicate that the color shift of the down-light LED luminaire induced by the optical property degradation of Microcellular PET aged at 85 ºC for 4000hours is 0.0001 and its lumen efficiency decreased by 1.57%. However, with the result of humidity test at 85 ºC and 85%RH, the application of this material is limited to the drying environment.

In the next step, in order to quantify the color shift effects induced by degradation of each individual part of the luminaire, diffuser and LED package will be involved in the thermal degradation, which will contribute to the development of the luminaire level color shift acceleration method.

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Chapter 5

Color Shift Acceleration on Mid-Power LED Packages[4]

Mid-power LED packages, whose electrical power rating ranges from 0.2 to 0.5W are now widely used in many indoor illumination applications and instead of other high cost device due to many advantages. The aim of this report is to investigate the color shift modes and mechanisms caused by different acceleration stresses and propose an acceleration and prediction method. Temperature stress, humidity stress and current stress were experimentally designed and performed to accelerate the color shift of mid-power LED packages and color shift mechanisms have been discussed based on the color shift results obtained from measurements. Conclusions could be drawn: 1） Exponential fitting demonstrates a good exponential relationship between color shift (Δu' Δv') and aging time almost for all the aging conditions. We can extrapolate the color shift Δu' and Δv' based on the fitted regression equations and then make the prediction for the total color shift Δu'v'； 2) Current stress can induce a different failure mode. Peak intensity reduction analysis reveals that the current stress accelerates the degradation of LED die; 3) Humidity test induced a substantial color shift both in u' and v'. Peak intensity comparison analysis reveals that not only the degradation of the LED die, but also degradation of the phosphor layer contributes to such high color shift in the humidity test.

[4] This chapter is derived from the submission: Guangjun Lu, W.D. van Driel, Xuejun Fan, Jiajie Fan, Cheng Qian, G.Q. Zhang, Color Shift Acceleration on Mid-Power LED Packages, Microelectronics Reliability, Accepted.

5.1 Introduction

Mid-power LED packages, whose electrical power rating ranges from 0.2 to 0.5W are currently widely used in many indoor illumination applications and instead of other high cost devices due to many advantages such as cost-effective, ease of installation because of simpler design for the distributed light system [1-3].

Some studies have been reported on the lumen output and degradation mechanisms of LED packages under different aging conditions [1-8]. There are also some studies on the color shift mechanisms. Meneghini and Zanoni et al investigated InGaN/GaN based high brightness LEDs with high temperature or dc or pulsed stress, results shown that thermal treatment can induce a worsening of the chromatic properties of the devices. In most of the cases, white LEDs submitted to stress tests show significant modifications of their spectral characteristics during stress time and, in particular, a decrease in the ratio between the intensity of phosphor-related emission and the intensity of the main blue peak. Consequently, after stress, the chromatic properties of the devices can shift toward a bluish light. The degradation of the chromatic properties is usually ascribed to the browning of the material used for the encapsulation of the phosphors/chip system. The degradation can be also correlated to the partial carbonization of the reflective surface of the package: this effect may introduce a decrease in the efficiency of the light extraction process due to the reduced contributions of the light reflected by the package to the overall emission. Both these processes can modify the spectral content of the light emitted by the LEDs and induce modifications in the relative intensity of the blue and yellow emission peaks. [9-14]

Huang et al studied the color shift caused by the yellowing of package encapsulate for mid-power white light LED packages in the outdoor illumination applications with high humidity and high temperature (WHTOL) [15]. Mehr et al experimentally investigated the color shift of remote phosphor plates made from Bisphenol-A polycarbonate (BPA-PC) and phosphor coating layer [16-19].

Davis et al [20-24] examined chromaticity shift modes of the PAR38 lamps with four types of built-in LED packages, results shown the chromaticity shift is dominated by the characteristics of the LED, since the optical materials changed

little in most samples. There are mainly four potential chromaticity-shift directions of a light source: blue shift, yellow shift, green shift and red shift.

However, research on the color shift and mechanisms of mid-power LED packages for indoor illumination applications, especially for the color shift acceleration and prediction, is less published. Since the color is important in some applications, although there is only one industry standard to define the color shift limit [25-27], it is important to predict the color shift for LED package designers and manufacturers.

The aim of this report is to investigate the color shift modes and mechanisms of mid-power LED packages caused by different acceleration stresses and propose an acceleration and prediction method for color maintenance. Temperature stress, humidity stress and current stress were experimentally designed and performed to accelerate the color shift of mid-power LED packages, and color shift mechanisms are discussed based on the color shift results obtained from measurements. A prediction method is proposed.

The experimental results and the proposed prediction method will be helpful in the mid-power LED package design and application, and also helpful in the color shift investigation and acceleration for the luminaire level products in which this class of LED packages is used.

The remainder of this paper is organized as follows: Section 2 presents the materials and methods for the experiment set-up and testing. Section 3 provides results and discussions on the designed experiment. Finally, concluding remarks are presented in Section 4.

5.2 Experiments and Methods

Samples used in the investigation are widely used mid-power LED packages, 5630 with dimension 5.6×3.0×0.96mm3, from a leading manufacturer in the industry. LED packages were mounted on metal core based plates as shown in Figure 1.

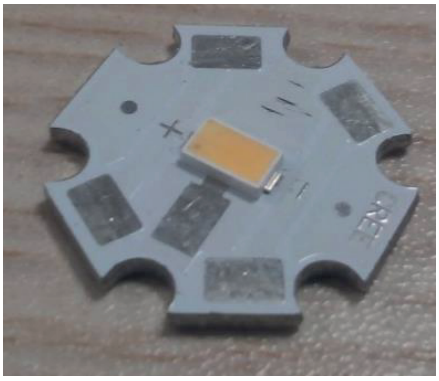


Figure 1 *LED package mounted on the metal core based plate*

Samples were divided into 3 different groups, and each group was subjected to a different aging condition, 25℃(room temperature), 85℃, or 85℃&85%RH. 25℃ is used here as a reference temperature to simulate the daily use condition, 85℃ is a temperature lift of 60℃ to investigate the temperature effect on color shift and 85℃&85%RH is for humidity test investigation. Each group has 2 sub-groups, which were powered on with a different current, 75mA or 225mA during aging. Lighting status is shown in Figure 2. Samples were taken out periodically and measured at room temperature with normal operation conditions by the same integrating sphere, as shown in Figure 3, with a diameter of 0.5m at 2pi mode.

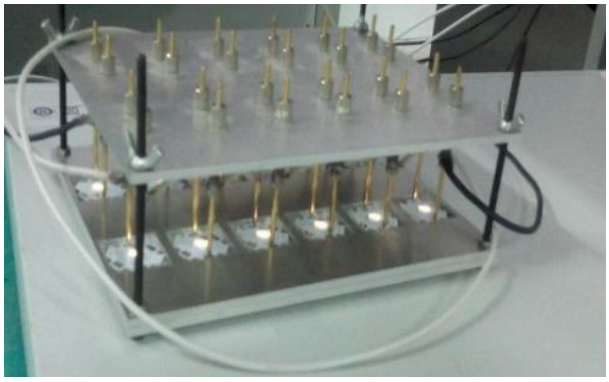


Figure 2 *LED packages lighted on during aging*

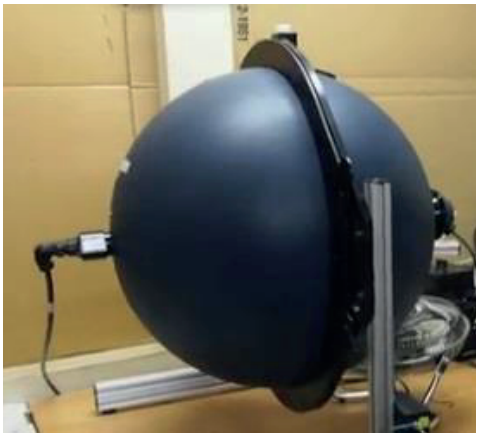


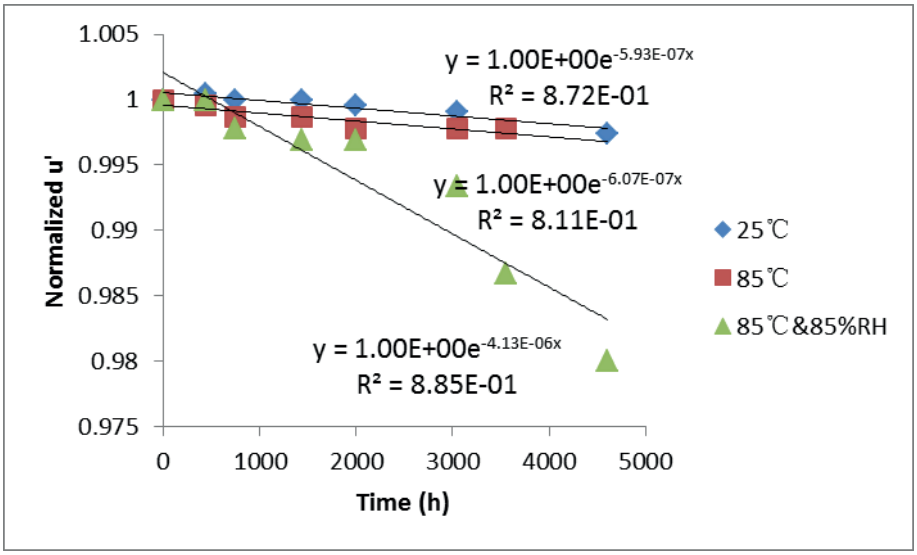
Figure 3 *Integrating sphere used for chromaticity measurement during aging*

5.3 Results and Discussion

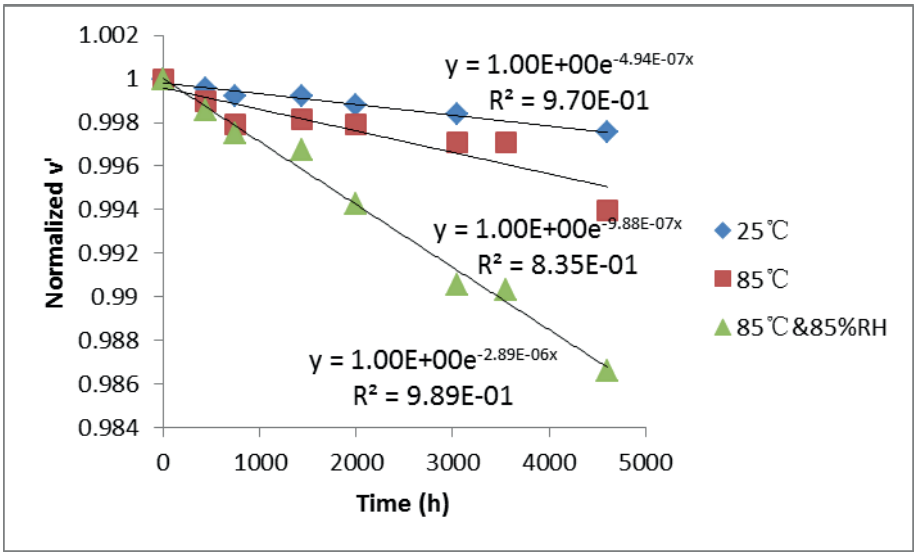
5.3.1 Results for the subgroup with loading current I= 75mA

Chromaticity coordinates data was collected, u' and v' (loading current I= 75mA) were normalized to their initial color points and shown in Figure 4 (a) and (b), respectively. Exponential model fitting demonstrates R2 (a scalar measure of model significance) is greater than 80% for all the aging conditions, which reveals a good exponential relationship between color shifts (Δu', Δv') and aging time. Negative reaction rate reveals a trend of decrease or degradation both in u' and v' under all the aging conditions.

Obviously, the aging condition of 85%RH&85℃ has a degradation rate greater than the other conditions in absolute value, which indicates humidity has a good acceleration effect in color shift. Similar phenomenon could be found in Figure 5, where samples were subjected to a greater loading current as high as 225mA during aging.



(a) Normalized u'

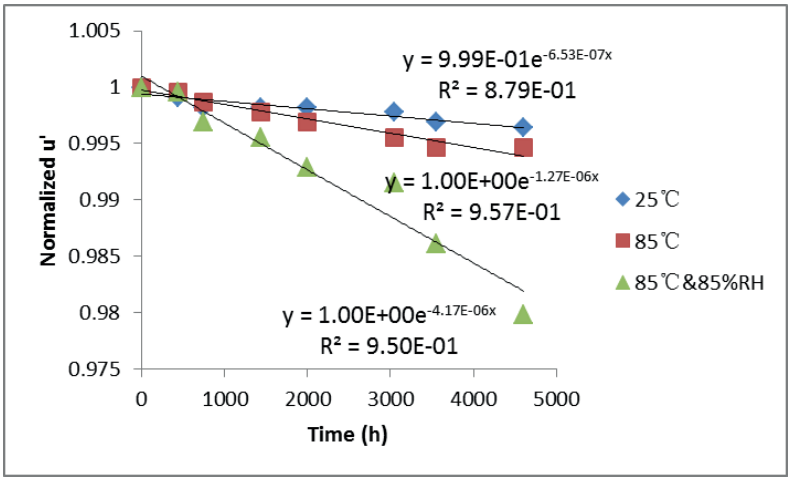


(b) Normalized v'

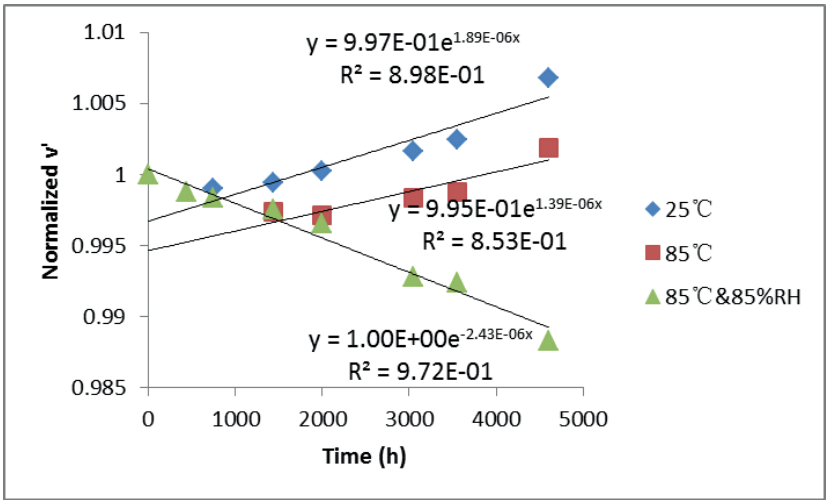
Figure 4 *Normalized u'and v' and exponential model fitting with different aging conditions (loading current I=75mA)*

5.3.2 Results for the subgroup with loading current I= 225mA

As mentioned, normalized u' and v' (aging current I= 225 mA) were shown in Figure 5 (a) and (b) respectively.



(a) Normalized u'



(b) Normalized v'

***Figure** 5 Normalized u'and v' and exponential model fitting with different aging conditions (loading current I=225mA)*

Similarly, exponential model fitting results demonstrate that R^2 is greater than 80% for all the aging conditions, which reveals again good exponential relationships between color shifts (Δu', Δv') and aging time.

Negative reaction rate reveals a trend of decrease in u' during aging under all conditions, while presences of positive reaction rate indicates a trend of increase in v' during aging at room temperature or 85 ℃, shown in Figure 5(b).

5.3.3 Discussions

To further understand this color shift phenomenon, SPDs have been checked for all the samples aged under different aging conditions. Both blue light peak intensity and yellow light peak intensity were gradually reduced during aging, but no obvious shift of peak wavelength was found, which is probably the reason that color shift (Δu', Δv') has an exponential relationship with duration of operation. An example , shown in Figure 6, demonstrates the relative flux intensity distributions of one sample aged at 85%RH&85℃ for different periods of time.

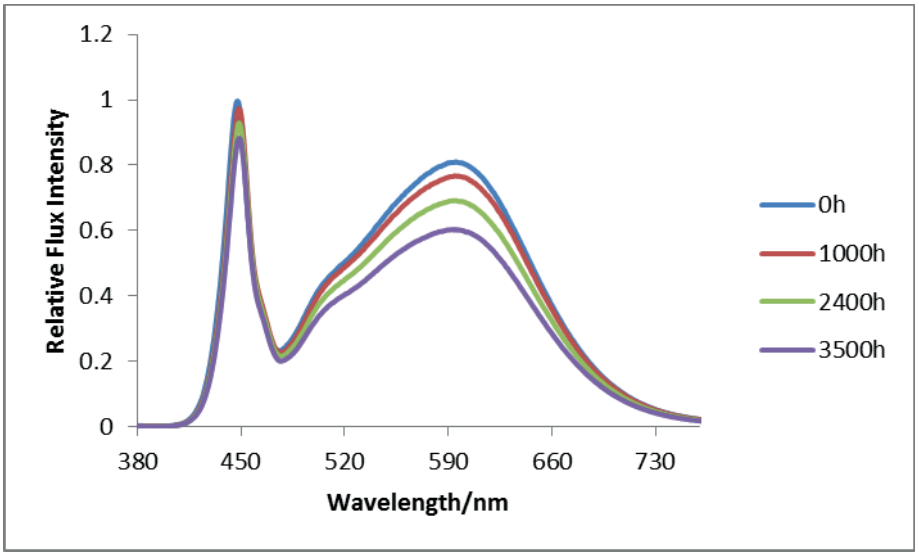
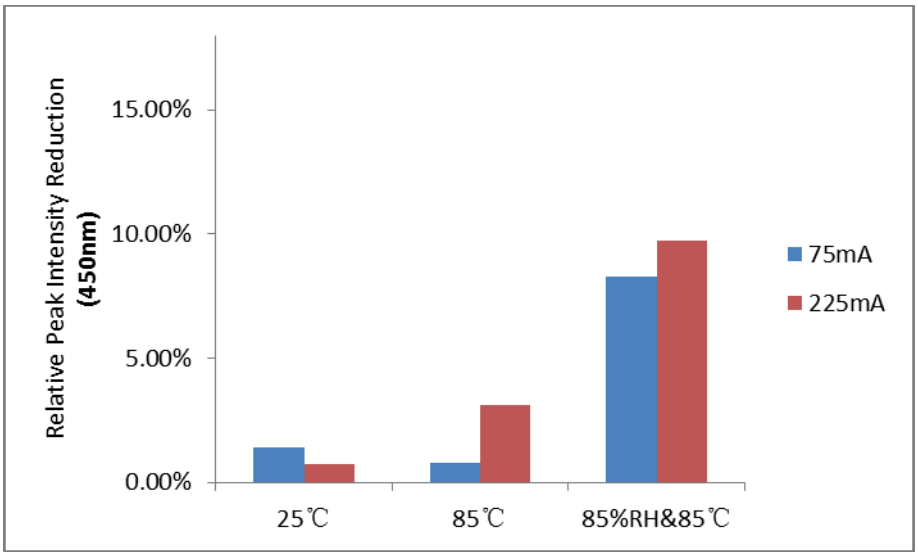
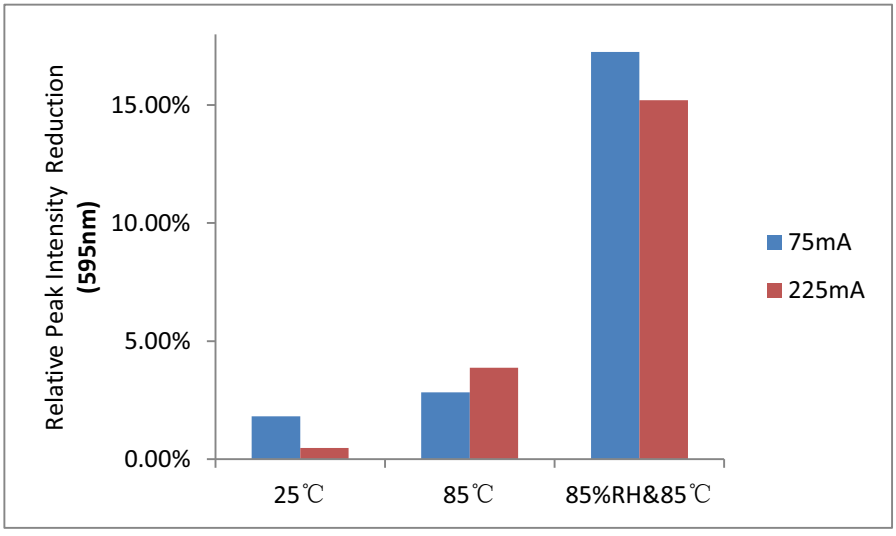


Figure 6 *SPDs of LED packages under different aging conditions*



(a) Blue light peak intensity reduction



(b) Yellow light peak intensity reduction

Figure 7 *Relative light peak intensity reduction after aging for 3500h with different conditions*

Since the color shift is caused by the change of the ratio of blue to yellow light, peak intensity reduction could be used to further investigate the color shift mechanisms with different aging conditions. Relative peak intensity reductions of blue light and yellow light for the samples after aging of 3500h are shown in Figure 7. After aging at 85℃ for 3500h, the reduction of blue peak intensity of the subgroup with high loading current (225mA) is several times higher than the other subgroup with low loading current (75mA), compared to a slight increase in the reduction of yellow peak intensity, thus leading to a different change of the ratio of blue to yellow light. That's why we observed a trend of increase in v' in Fig 5 (b), compared to a decrease in Figure 4(b). The blue light peak reduction during aging at 85℃ is mainly associated with the degradation of the LED die. The junction temperature will be increased with the stress current. Both the increased stress current and the lifted junction temperature will accelerate the degradation of the LED die and hence induce the increase of blue light peak intensity reduction. Another factor is the conversion efficiency of phosphor. The lifted temperature by stress current will also worsen the conversion efficiency and accelerate the reduction of the yellow light peak intensity.

There seems to be a different phenomenon for the samples aged at room temperature. When the stress current is increased to 225mA during aging, the blue light peak intensity reduction becomes less. That's probably because the acceleration of current is not significant at low temperature and less blue light conversed to the yellow light during aging, and the phosphor conversion efficiency has not been worsen, or even become better. Subgroup to subgroup variation is also a factor.

Obviously, after humidity stress test, both the blue peak intensity and the yellow peak intensity have been reduced substantially for both stress currents. That's why we observed a rapid decrease in u' and v' in Figure 4 and 5. The higher stress current has a more significant reduction in blue light peak intensity, which is consistent with the observation of aging at 85℃. The humidity stress accelerates the reduction of blue peak intensity, as reported earlier in Huang and Tan's works [3][25], the moisture has been penetrated deep into the phosphor layer, and normal operation is not likely to drive out the moisture, or a blue light over-absorption was induced , which could probably

cause a localized temperature lift as high as 300℃. This lifted temperature is so high that will not only accelerate the degradation of the LED die, but also worsen the phosphor layer including the conversion efficiency of phosphor particle and importantly, the transmittance change of phosphor plate. That's why the yellow light peak intensity has been reduced even more as compared to the blue light peak intensity.

Since differences exist between the mechanisms of color shift under different stress conditions, we should be careful when we choose an acceleration method. For example, when the LED package is used in the dry environment, if the humidity test is used as an acceleration stress, it will induce a substantial reduction in color shift since new factor such as transmittance change has been involved. In the normal operation, when the current is used as an acceleration factor, it will lead to a different color shift trend in v'. Nevertheless, the acceleration is possible if we can find the correlation of acceleration between different stresses.

Color shift is complex, what we care is more about the prediction. Since there is likely an exponential relationship between color shift (Δu' and Δv') and duration of operation, we can extrapolate the color shift Δu' and Δv' based on the fitted regression equation and then make the prediction for the total color shift Δu'v'.

5.4 Conclusions

Temperature stress, humidity stress and current stress were experimentally designed and performed to accelerate the color shift of mid-power LED packages and color shift mechanisms have been discussed based on the color shift results obtained from measurements. Conclusions could be drawn as below:

1) Exponential fitting demonstrates a good exponential relationship between color shift (Δu', Δv') and aging time almost for all the aging conditions. According to the investigation of SPDs, both blue light peak intensity and yellow light peak intensity gradually decreased during aging, but no obvious shift of peak wavelength was found, which probably is why the exponential relationship exists. We can extrapolate

the color shift Δu' and Δv' based on the fitted regression equations and then make the prediction for the total color shift Δu'v'.

2) Negative reaction rate reveals a trend of decrease in u' during aging under all conditions, while presences of the positive slope indicates a trend of increase in v' during aging at room temperature or 85 ℃ when the stress current is lifted to 225mA, which indicates the current stress can induce a different failure mode. Peak intensity reduction analysis reveals that the current stress accelerates the degradation of LED die.

3) Humidity test induced a substantial color shift both in u' and v'. Peak intensity comparison analysis reveals that not only the degradation of LED die, but also degradation of the phosphor layer including the conversion efficiency of phosphor particle and more importantly, the possible transmittance change of phosphor plate contributes to such high color shift.

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Chapter 6

A Novel Approach of Color Shift Investigation on LED-based Luminaires[5]

This paper describes a novel approach to the investigation of the color shift of LED-based luminaires. This approach is based on a view factor method and can easily be used to extract the color shift contribution from each individual component of the LED luminaires. According to the results from the simulations and calculations, after aging at 85 °C for 4000 h, the LED downlight exhibited a total color shift of about 0.002, which is less than that of the LED packages of around 0.0025. This is because the two major contributors, the LED package and diffuser, oppositely contribute to the color shift component of $\Delta v'$. The $\Delta v'$ component of the LED package decreases by 0.0026 compared to an increase of about 0.0017 caused by the change in diffuser transmission during aging. The color shift induced by the aging of LED packages is quite different from that of the diffuser. The relative flux degradation of the LED packages in the spectral power distribution clearly occurs in the yellow light area, which causes the color to shift to blue. In contrast, the color will shift to yellow for the aging of the diffuser as there is a greater degradation of flux in the blue light area. The results obtained from the measurement and from the proposed approach are comparable.

[5] This chapter is derived from the submission: Guangjun Lu, W.D. van Driel, Xuejun Fan, Jiajie Fan, Cheng Qian, Huaiyu Ye, G.Q. Zhang, A Novel Approach of Color Shift Investigation on LED-based Luminaires, under Journal review

6.1 Introduction

Solid state lighting products are increasingly being used worldwide for general lighting because of the continuous remarkable progress made in lighting efficacy, cost reduction, and quality improvement, thus gaining recognition from consumers. Unlike traditional lighting, the luminous flux maintenance and color stability are two important factors for evaluating long-term reliability, which is always a concern for some potential consumers and hence prevents their use to a certain extent [1]. The DOE Energy Star Program requires that the change in chromaticity over the minimum lumen maintenance test period (6000 hours) should be within 0.007 on the CIE 1976 (u', v') diagram [2]. Poor color maintenance can be a substantial problem in applications where color quality is important, including museum and gallery lighting, architectural facade lighting, retail display lighting, healthcare lighting, hospitality applications, cove and wall wash lighting, and down lighting in commercial and residential applications [3].

Compared to traditional incandescent lighting products, the color shift mechanisms of LED lighting are more complex because they consist of several different components, each of them having the potential to limit the long-term stability and contribute differently to the color shift during operation. Some studies have been conducted on the color shift effects and mechanisms of LED packages, reflectors, and diffusers [4–21].

The factors impacting the color point stability of LEDs include aging-induced changes in the phosphor, emitter, and encapsulate materials. Emitters can exhibit a decrease in radiant flux over time; phosphors can experience decreases in quantum efficiency or shifts in the emission spectrum caused by oxidation; encapsulates can exhibit cracking, oxidation and yellowing, or changes in the index of refraction. Higher temperatures will accelerate these degradation mechanisms, leading to a greater color shift, but the magnitude of the color shift as a function of temperature will vary with packaging material and manufacturing process. The resulting direction of color shift depends on the dominant degradation mechanisms occurring in the package, which, in turn, depends on the packaging material and method of construction [22–24].

Davis et al. examined the chromaticity shift modes of PAR38 lamps with four types of built-in LED packages, and the results showed that the

chromaticity shift was dominated by the characteristics of the LED as the optical materials changed little in the majority of samples. There are four main potential chromaticity shift directions of a light source: blue shift, yellow shift, green shift, and red shift [25–29].

Color shift acceleration models are required to verify the prediction method, especially for LED-based luminaires. The reliability of such calculations is a present concern, although the Illuminating Engineering Society has initiated a committee tasked with developing an approved procedure. This work is targeted toward LED packages, for which the exact operating characteristics are more easily controlled. Similar to lumen maintenance and reliability, extending the component-level data to complete LED lamps and luminaires can be difficult, and many challenges exist [27]. A lack of standard procedures for predicting color stability performance has contributed to user uncertainty—potentially limiting adoption and making it more challenging for manufacturers to provide warranty coverage for the color shift [3]. This paper proposes a view factor approach for the color shift investigation of LED-based luminaires. It can be easily used to extract the color shift contribution of each individual component of LED luminaires, which will pave the way toward future color shift acceleration and prediction.

6.2 Materials and Methods

In order to develop the color shift acceleration and prediction method, it is important to investigate the color shift contribution from each individual component. Hence, a novel approach for color shift investigation based on a view factor method is proposed in this section. The downlight to be investigated was aged at 85 ℃ in an oven and was switched on for around 4000 h. The soldering temperature was measured with a thermal couple to be about 105 ℃. To quantify the color and lumen change of the LED packages mounted in the downlight during aging, the same type of LED package from the same supplier was separately mounted, placed in the same oven, and switched on with an equivalent current. The soldering temperature of the LED package was measured to also be around 105 ℃. The light flux, color, and spectral power distributions (SPDs) of those LED packages were measured and recorded by an integrating

sphere before and after aging. The diffuser used in the downlight was a type of commercial misty PMMA with a thickness of 2 mm. The transmittances of the diffuser before and after aging were measured. The reflective material used in the downlight was a type of commercial PET, whose reflectivity before and after aging was also measured. Refer to [20] for more details.

To help illustrate this approach, a schematic of light paths for the investigated downlight is shown in Figure 1. Each light path exchange can be considered a contribution to the color shift. Equation (1) can be used to describe the exchange.

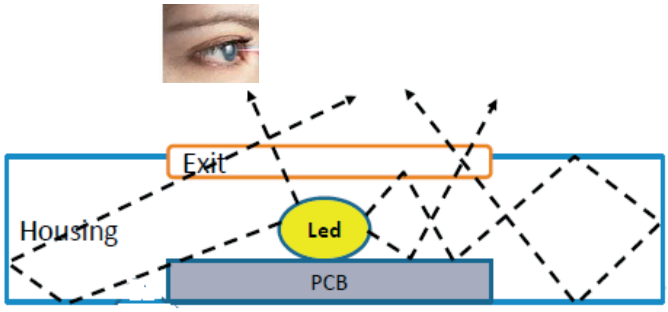


Figure 1 *Schematic of light paths*

$$\begin{pmatrix} \Phi_{itoLEDs} \\ \Phi_{itopcb} \\ \Phi_{itohou\sin g} \\ \Phi_{itoexit} \end{pmatrix} = \begin{pmatrix} F_{L-L} & F_{P-L} & F_{h-L} & F_{e-L} \\ F_{L-P} & F_{P-L} & F_{h-p} & F_{e-p} \\ F_{L-h} & F_{P-h} & F_{h-h} & F_{e-h} \\ F_{L-e} & F_{P-e} & F_{h-e} & F_{e-e} \end{pmatrix} \begin{pmatrix} \Phi_{ifromLEDs} \\ \Phi_{ifrompcb} \\ \Phi_{ifromhou\sin g} \\ \Phi_{ifromexit} \end{pmatrix} \tag{1}$$

Where Φ represents the light flux and *i* denotes the number of reflections made. *F* is the so-called view factor (conservation of light: sum of each matrix column = 1). The Light Tools software was used to extract the view factor *F* and light flux Φ. The physics of reflection can be described by equation (2).

$$\begin{pmatrix} \Phi_{i+1\,fromLEDs} \\ \Phi_{i+1\,frompcb} \\ \Phi_{i+1\,fromhou\sin g} \\ \Phi_{i+1\,fromexit} \end{pmatrix} = \begin{pmatrix} R_L & & & \\ & R_p & & \\ & & R_h & \\ & & & R_e \end{pmatrix} \begin{pmatrix} \Phi_{itoLEDs} \\ \Phi_{itopcb} \\ \Phi_{itohou\sin g} \\ \Phi_{itoexit} \end{pmatrix} \tag{2}$$

Each reflection is accompanied by a small change in color, $\Delta u_i'$ and $\Delta v_i'$,

depending on the *L* (LED), *p* (PCB), *h* (housing), and *e* (exit) components. The exit transmission can be described by equation (3). It is also accompanied by a small change in color during transmission. Φtoexit is the light flux toward the exit before transmission, which can be calculated via equation (4).

$$\Phi_{fromexit} = (1 - R_e - A)\Phi_{toexit} \tag{3}$$

$$\begin{aligned} \Phi_{toexit} &= \sum_i \Phi_{itoexit} \\ &= F_{L-e}\sum_i \Phi_{ifromLEDs} + F_{h-e}\sum_i \Phi_{ifromhousing} + F_{p-e}\sum_i \Phi_{ifrompcb} + F_{e-e}\sum_i \Phi_{ifromexit} \end{aligned} \tag{4}$$

The color shift $\Delta u'$, $\Delta v'$, and $\Delta u'v'$ of the flux to the exit Φ_{toexit} in the downlight can be estimated via equations (5–7). $\Delta u_L'$, $\Delta u_p'$, $\Delta u_h'$, and $\Delta u_e'$ denote the color changes of the fluxes exchanged on the *L*, *p*, *h*, and *e* components. *r* is the ratio of the flux returned to the exit after a cycle of reflections in the downlight. In addition, they can all be obtained via Monte Carlo simulations with Light Tools. The change in color caused by the transmission of the exit diffuser can also be obtained via a Monte Carlo simulation.

$$\Delta u' = w_{LED} \bullet \Delta u_L' + w_{pcb} \bullet \Delta u_P' + w_{housing} \bullet \Delta u_h' + w_{housing} \bullet \Delta u_e' \tag{5}$$

$$\Delta v' = w_{LED} \bullet \Delta v_L' + w_{pcb} \bullet \Delta v_P' + w_{housing} \bullet \Delta v_h' + w_{housing} \bullet \Delta v_e' \tag{6}$$

$$\Delta u'v' = \sqrt{\Delta u'^2 + \Delta v'^2} \tag{7}$$

Where w_{LED}, w_{PCB}, $w_{housing}$, and w_{exit} can be described as follows:

$$w_{LED} = \frac{F_{L-e}\sum_i \Phi_{ifromLEDs}}{\Phi_{toextt}}$$

$$w_{pcb} = \frac{F_{p-e}\sum_i \Phi_{ifrompcb}}{\Phi_{toexit}}$$

$$w_{housing} = \frac{F_{h-e}\sum_i \Phi_{ifromhousing}}{\Phi_{toexit}}$$

$$w_{exit} = \frac{r\sum_{i-1} \Phi_{ifromexit}}{\Phi_{toexit}}$$

To perform the simulations for extraction, the initial condition is shown in equation (8).

$$\begin{pmatrix} \Phi_{0\,fromLEDs} \\ \Phi_{0\,frompcb} \\ \Phi_{0\,fromhou\sin g} \\ \Phi_{0\,fromexit} \end{pmatrix} = \begin{pmatrix} \Phi_{LEDs} \\ 0 \\ 0 \\ 0 \end{pmatrix} \tag{8}$$

The following assumptions were made: the LED is top-emitting; light exits from the LEDs; no light bounces from the LEDs.

6.3 Results and Discussions

6.3.1 Measured results for color shift of downlights after aging

The SPD, lumen, and color data of the downlight before and after aging were measured and recorded by an integrating sphere with a diameter of 2 m at the 2π mode. Figure 2 presents the SPD of the downlight before and after aging. A clear degradation of flux intensity can be observed both in the yellow light area (around 590 nm) and blue light area (around 450 nm). The changes in lumen and color of this downlight are presented in Table 1.

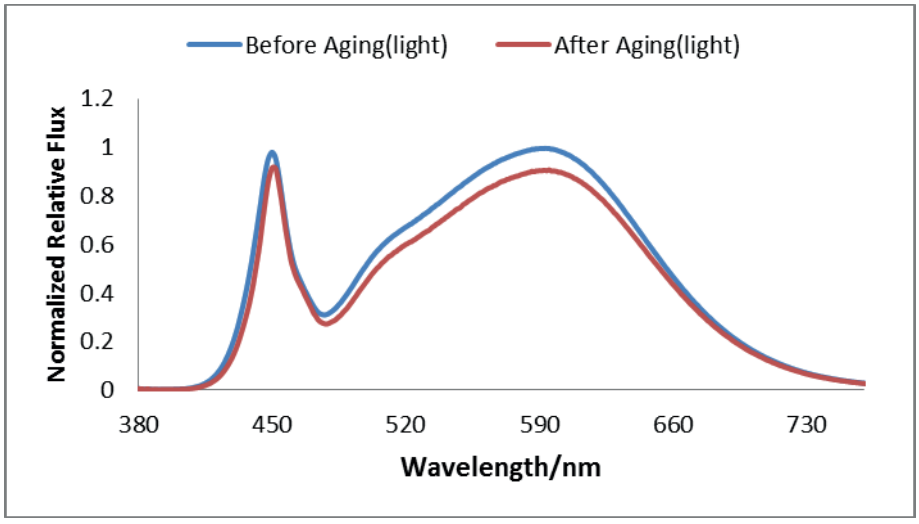


Figure 2 *SPDs of downlight before and after aging*

Table 1 Lumen and color data measurement before and after aging

			Before aging	After aging
Power		(W)	10.1	10.1
Luminous Flux		[lm]	936.65	838.66
Lumen Degradation		[-]	10.46%	
Color point	u'	[-]	0.2229	0.2248
	v'	[-]	0.5026	0.5041
Color shift	Δu'	[-]	0.0019	
	Δv'	[-]	0.0015	
	Δu'v'	[-]	0.0024	

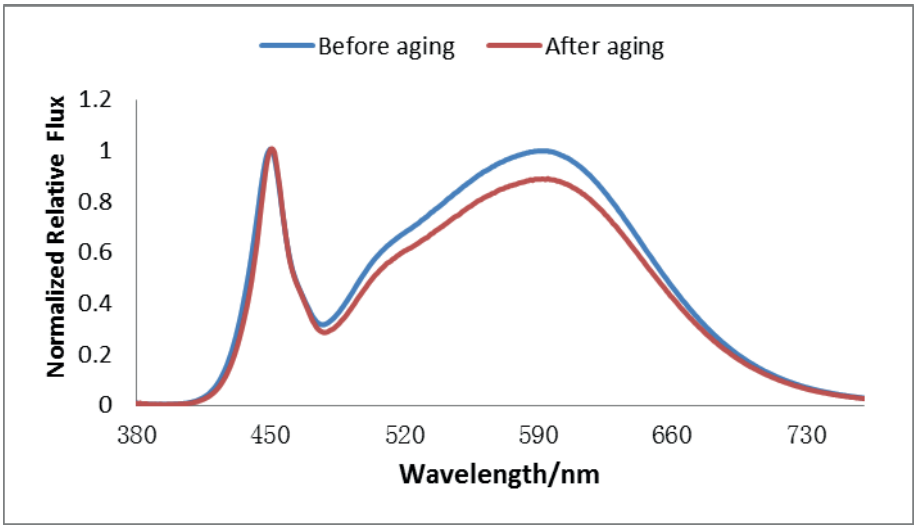


Figure 3 *SPDs of LED packages before and after aging.*

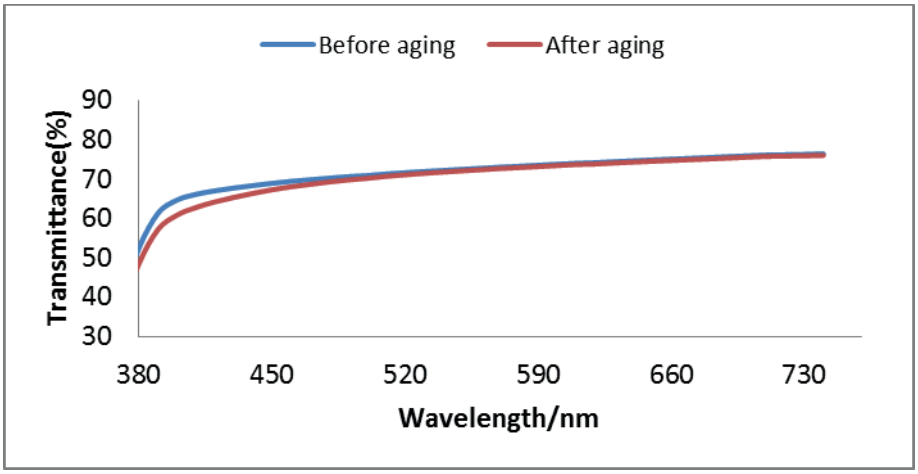


Figure 4 *Measured transmittance of PMMA diffuser before and after aging.*

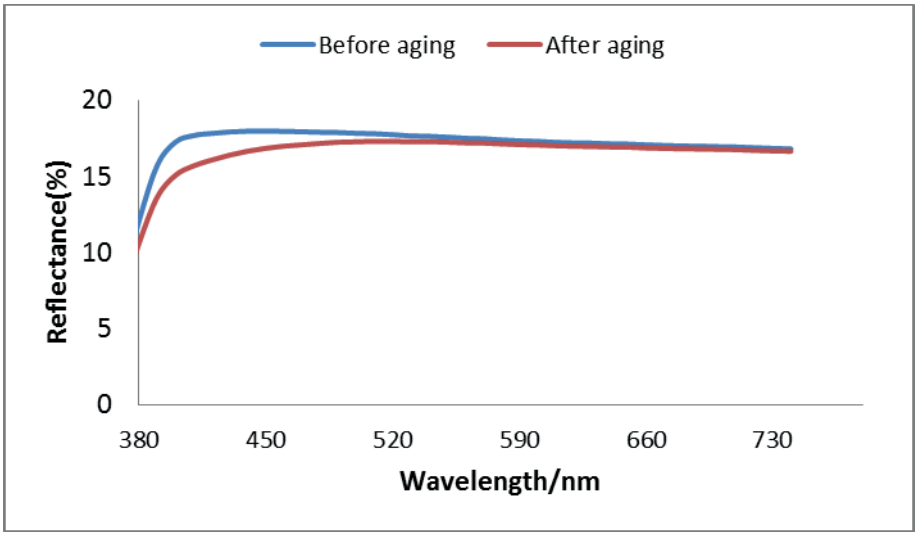


Figure 5 *Measured reflectance of PMMA diffuser before and after aging*

6. 3.2 Inputs for simulation

The average SPDs of the LED packages before and after aging are presented in Figure 3, which will be used as inputs for the simulation.

Both the transmittance and reflectance of the diffuser before and after aging were measured, as shown in Figures 4 and 5, respectively, which will be used as

inputs for the simulation.

The PET was not only mounted on the sidewall, which was used as the housing reflective material, but also covered the PCB surface. Thus, the housing and contribution of the PCB to the color shift depends on the change in reflectance of the PET. Figure 6 presents the measured reflectance before and after aging, which will be used as inputs for the simulation.

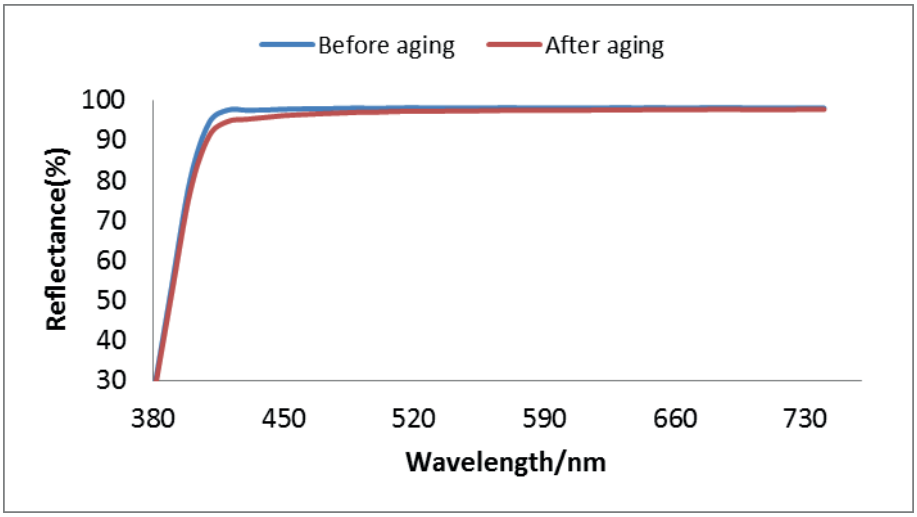


Figure 6 *Measured reflectance of PET material before and after aging*

6.3.3 Color shift results

1) Color shift before aging (0 h)

Table 2 View factors at 0 h

t=0 h		From			
		LEDs	PCB	Housing	Exit
To	LEDs	0.000	0.000	0.000	0.000
	PCB	0.000	0.000	0.245	0.534
	Housing	0.214	0.214	0.000	0.466
	Exit	0.786	0.786	0.755	0.000

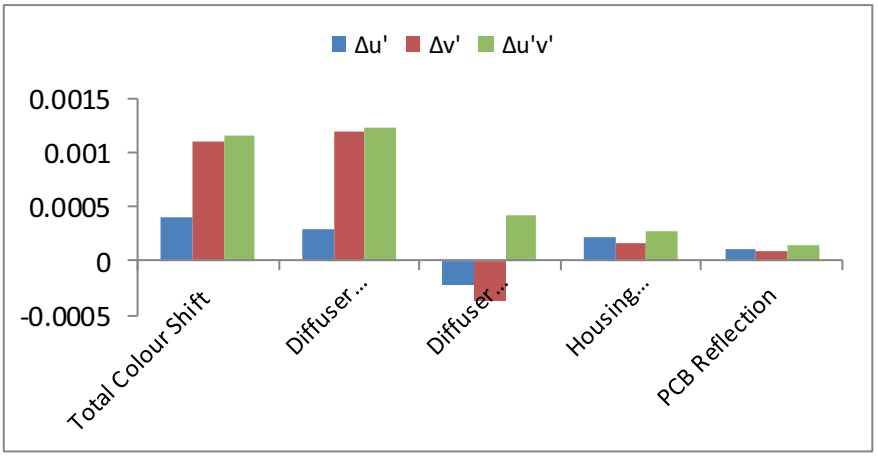


Figure 7 *Color shift of downlight at 0 h*

The view factors for the downlight before aging were extracted by a Monte Carlo simulation, as shown in Table 1. The color shift of each individual component was also obtained via simulations. Accordingly, the color shift of the downlight was calculated. Figure 7 presents the color shift of the downlight at 0 h and the contribution from each individual component. Even for the non-aged case (at 0h), the light coming from the downlight exhibits a color change of approximately 0.001 compared with the color of the light emitted from the LED packages. It can be observed that the diffuser is a major contributor. When the light passes through the diffuser, the ratio of blue light to yellow light in the SPD changes because of the transmittance difference between wavelengths, which leads to the change in color [16][18].

2) Color shift after aging (4000 h)

Table 3 View factors at 4000 h

t=4000h		From			
		LEDs	PCB	Housing	Exit
To	LEDs	0.000	0.000	0.000	0.000
	PCB	0.000	0.000	0.239	0.536

	Housing	0.214	0.214	0.000	0.464
	Exit	0.786	0.786	0.761	0.000

Similarly, we obtained the view factors (presented in Table 3) for the downlight after aging for 4000 h. The color shift of each individual component and, accordingly, that of the downlight were calculated. Figure 8 presents the color shift of the downlight after aging for 4000 h and the contribution from each individual component.

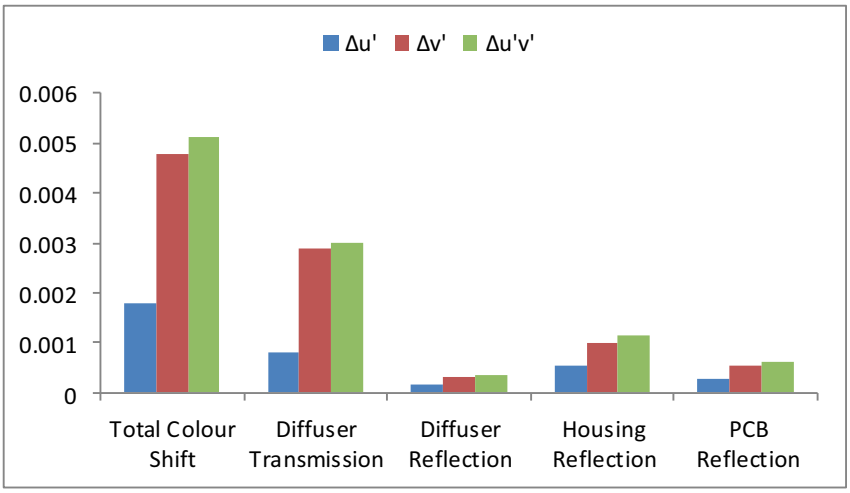


Figure 8 *Color shift of downlight at 4000 h*

6.3.4 Comparison and discussion

Note that the color changes shown in Figures 7 and 8 do not include the color shift of the LED packages. Figure 9 presents the color shift and contribution of each individual component including the LED packages caused by aging.

According to the results from the simulation and calculation method, after aging for 4000 h, the downlight exhibits a total color shift of about 0.002, which is less than that of the LED packages of around 0.0025. This is because the two major contributors, the LED package and diffuser, oppositely contribute to the color shift component of $\Delta v'$. The $\Delta v'$ component of the LED package decreases by 0.0026 compared to an increase of about 0.0017 caused by the change in diffuser transmission during aging. The color shift induced by the aging of LED

packages is quite different from that of the diffuser. The relative flux degradation of the LED packages in the SPD clearly occurs in the yellow light area, which induces the color to shift to blue. By contrast, the color will shift to yellow for the aging of the diffuser as there is much more degradation of flux in the blue light area.

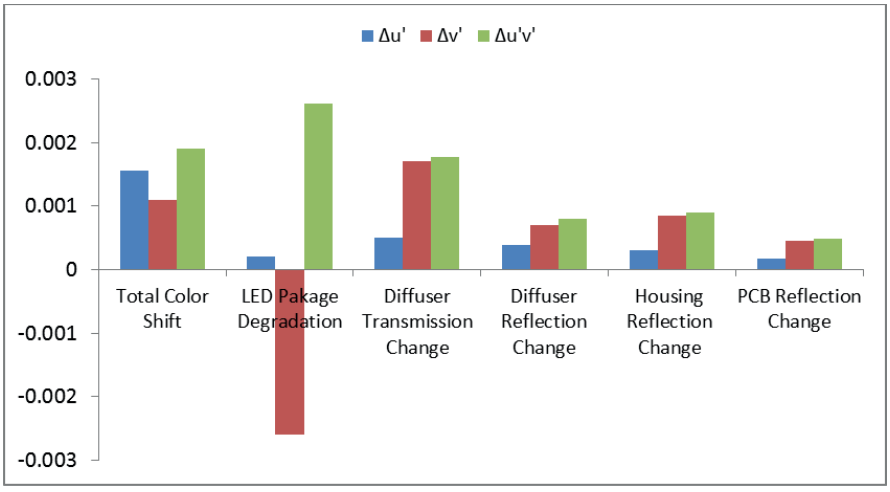


Figure 9 *Color shift contribution after aging*

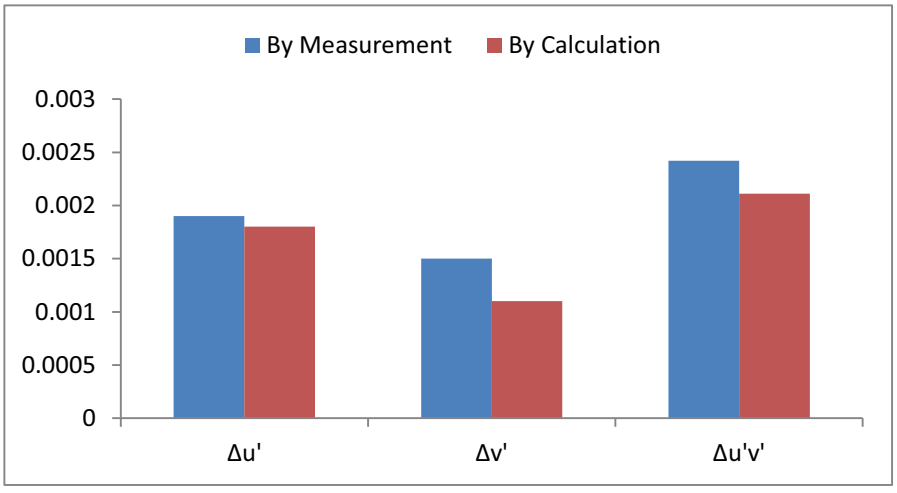


Figure 10 *Color shift comparison between measurement and proposed approach.*

Figure 10 presents the comparison between the measurement and proposed approach. It can be observed that the results are comparable. The errors for the approach proposed in this section can be attributed to several factors. One is the assumption that there is no light bouncing between the LED packages, which would slightly decrease the color change. The other factor is the measurement error of the optical parameters as inputs for the simulation.

6.3.5 Luminaire color shift acceleration and prediction proposal

Based on the above investigation, we can separate the total color shift $\Delta u'v'$ into the components $\Delta u'$ and $\Delta v'$ for acceleration and prediction. As each individual component has a different contribution and mechanism, we can add the contribution from each individual component to the color shift component, and the acceleration can be performed individually. If we only consider the temperature aging for acceleration, the prediction can be described by equations (9–11).

$$\Delta u'(t,T) = \Delta u'_{led}(t,T) + \Delta u'_{diffuser}(t,T) + \Delta u'_{reflector}(t,T) \tag{9}$$

$$\Delta v'(t,T) = \Delta v'_{led}(t,T) + \Delta v'_{diffuser}(t,T) + \Delta v'_{reflector}(t,T) \tag{10}$$

$$\Delta u'v'(t,T) = \sqrt{\Delta u'^2(t,T) + \Delta u'^2(t,T)} \tag{11}$$

We can follow the steps shown below for the luminaire color shift acceleration and prediction.

Step 1: For the LED packages, determine the SPDs as F (time, temperature…) by integrating sphere measurement. For the other components, determine the optical parameters as F (time, temperature…) by optical measurement.

Step 2: Using step 1 as inputs, determine Δu' and Δv' as F (time, temperature…) for each component by simulation. Meanwhile, determine the view factor matrix as F (time, temperature).

Step 3: For each component, fit the acceleration model (linearity, Arrhenius + Weibul / lognormal, etc.).

Step 4: Calculate the system-level color shift based on equations (9–11).

6.4 Conclusions

This paper describes a novel approach for investigating the LED-based luminaire color shift. The following conclusions can be drawn after the investigation of an LED downlight before and after aging at 85 ℃ for around 4000 h.

- The approach proposed in this paper can easily be used to extract the color shift contribution of each individual component of LED luminaires.
- According to the results from the simulation and calculation method, after aging for 4000 h, the downlight exhibits a total color shift of approximately 0.002, which is less than that of the LED packages of around 0.0025. This is because the two major contributors, the LED package and diffuser, oppositely contribute to the color shift component of $\Delta v'$. The $\Delta v'$ component of the LED package decreases by 0.0026 compared to an increase of about 0.0017 caused by the change in diffuser transmission during aging.
- The color shift induced by the aging of LED packages is quite different from that of the diffuser. The relative flux degradation of the LED packages in the SPD clearly occurs in the yellow light area, which causes the color to shift to blue. By contrast, the color will shift to yellow for the aging of the diffuser as there is much more degradation of flux in the blue light area.

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Chapter 7

LED Based Luminaire Color Shift Acceleration and Prediction[6]

Light-emitting diode (LED) lighting generally comprises several different components, with a different color shifts occurring in each individual component via various mechanisms during the period of use. This paper proposes a method for predicting and accelerating color shift of LED-based luminaires; the proposed method is based on the view-factor approach. Based on the results of this investigation, several conclusions can be drawn. 1) The color shift induced by aging of LED packages is quite different from that obtained using diffusers and reflectors; aging causes a blue shift in LED packages, whereas reflectors and diffusers cause a yellow shift. 2) The overall color shift of LED-based luminaires is toward blue, and the direction of this shift is dominated by the LED packages. 3) The color shift caused by diffusers and reflectors cancels that caused by LED packages; therefore, the net color shift of a luminaire is lower than that of LED packages. 4) The proposed method may be used to predict color shift of LED-based luminaires.

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[6] This chapter is derived from the submission: Guangjun Lu, W.D. van Driel, Xuejun Fan, Jiajie Fan, Cheng Qian, G.Q. Zhang, LED-based luminaire color shift acceleration and prediction, under Journal review

7.1 Introduction

With increasing adoption of light-emitting diode (LED) lighting products and their use for extended durations, the problem of color shift LED products is becoming more apparent, especially in some important application areas such as museums and galleries, architectural facade, retail displays, healthcare, hospitality, cove and wall wash lighting, and down lighting in commercial and residential areas [1].

Several studies have reported the lumen output and degradation mechanisms responsible for aging under different conditions [2-5]. However, the mechanisms of color shift, and particularly methods for prediction and acceleration of color shift are not understood thoroughly.

On the basis of the CIE 1976 (u', v') diagram, the DOE Energy Star Program requires that the change in chromaticity over the minimum lumen maintenance test period (6000 h) shall be within 0.007 [6].

Some studies have investigated color shift mechanisms and their effects on LED packages, reflectors, and diffusers [7-21]. Davis et al examined the chromaticity shift modes of PAR38 lamps using four different types of built-in LED packages. The results show that the chromaticity shift is dominated by the characteristics of the LED because the optical materials changed only slightly in most samples. The chromaticity of a light source generally shifts in four directions, namely blue, yellow, green, and red. A blue shift may result from an increase in blue emission or a decrease in yellow emission; this is accompanied by a decrease in the chromaticity in both u' and v'. This type of color shift can be attributed to a loss of phosphor quantum efficiency due to chemical changes, raised temperatures, oxidation of the molding compound in a PLCC, operating the phosphor above the saturation flux level, settling or precipitation of the phosphor, or top-to-bottom fractures in the binder of the phosphor-binder layer, each resulting in blue photons bypassing the phosphor layer. A yellow shift may result from an increase in yellow emission or a decrease in blue emission; this is accompanied by an increase in the chromaticity in both u' and v'. This type of color shift can be caused by an increase in the phosphor quantum efficiency owing to chemical changes, temperature decreases, cracking or delamination of the phosphor-binder layer, or discoloration or via oxidation of the lenses or the

reflector. A green shift occurs when the chromaticity change deviates from the blue–yellow line toward the green direction; this is accompanied by a decrease in the chromaticity in u' and a minimal change and a possible increase in v' in some cases. A green shift is indicative of a change in the emission properties of either the blue or yellow emitter; this is attributed to the oxidation of the phosphor. A red shift occurs when the chromaticity change deviates from the blue–yellow line toward the red direction, accompanied by an increase in the chromaticity in u' and a minimal change in v'. A red shift is indicative of a change in the emission properties of the blue, yellow, or direct red emitters. [22-26]

Lall et al [27] used the exponential model shown in Equation (1) to predict the correlated color temperature (CCT) and lumen maintenance for LED lamps as well as for the LEDs inside the LED lamps when subjected to a 85°C, 85% RH test.

$$\Phi = \beta e^{\alpha t} \tag{1}$$

where β is the pre-decay factor, α is the decay rate, t is the test time, and Φ is the CCT or lumen maintenance depending on the decay rate being calculated. The decay rate is a function of temperature, as represented by Equation (2), which is also known as the Arrhenius equation.

$$\alpha = Ae^{-\frac{Ea}{K_b T}} \tag{2}$$

Where T is the temperature in kelvin, K_b is the Boltzmann constant, and E_a is the activation energy. The method of least squares (LS) was used to compute the decay rate for both CCT and lumen maintenance.

Mehr et al generalized the Eyring equation-reliability model, which is represented by Equation (3), to predict the lumen maintenance and CCT for remote phosphor plates subjected to a HAST test, which examines the effects of both light intensity and temperature.

$$R = \gamma_0 (I)^n e^{-\frac{Ea}{K_b T}} \tag{3}$$

where R is the reaction rate, γ_0 is the pre-exponential factor, I is the intensity of blue light, n is a constant, E_a is the activation energy, K_b is the Boltzmann

constant, and T is the absolute temperature. [28]

In addition, Koh et al demonstrated that a linear model derived by analysing both experiment and simulation data can be applied for introducing color shifts in LED devices [29]. Huang used this linear model to predict the color shift of mid-power white-light LED packages; in addition, the decay rate constant was estimated on the basis of the modified Wiener process. However, an analysis of early degradation was excluded [30].

To date, no method or approach has been widely accepted for predicting and accelerating color shift of LED lighting. This is mainly due to the high complexity of color shifts in LED lighting, which generally comprises several different components. Each one of these components contributes differently to the color shift during the period of use, both directly and by altering the physics of light emission during the period of application.

Here, we propose a novel method for the prediction and acceleration of color shifts in LED-based luminaires.

7.2 Materials and Methods

A downlight, shown in Figure 1, was used as a representative LED lighting device for this investigation. As shown in Figure 2, the LED packages, diffuser, and reflector each contribute to the color shift of the downlight.



Figure 1 *Downlight investigated herein for color shift prediction*

To quantify the color shift caused by each part, a temperature–acceleration technique was implemented on each component independently to simulate different aging conditions. Several samples of a LED package obtained from a supplier were separately mounted and exposed to the room temperature, 55℃, or 80℃. The Ts measured by a thermocouple was approximately 62℃, 75℃, and 85℃ under each of these conditions, respectively. Reflectors made of a commercial material known as MCPOLYCA were used in aging tests at temperatures below the Tg (approximately 105℃), i.e., at 55℃, 85℃, and 100℃. The diffuser used in the downlight and tested in this experiment was a commercially available misty PMMA with a thickness of 2 mm. Diffuser samples were exposed to aging temperatures below the Tg (approximately 120℃), i.e., 85℃, 100℃, and 115℃.

The light flux, color, and spectral power distributions (SPDs) of the LED packages were measured periodically over the course of temperature aging by using an integrating sphere. The transmittance of the diffuser and the reflectance of the reflector were also measured periodically during the aging period.

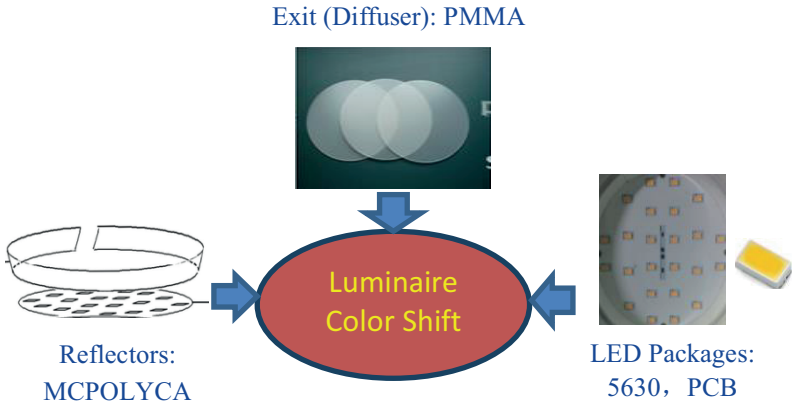


Figure 2 *Components of the downlight contributing to the color shift*

The downlight to be investigated was powered on in a room. The average recorded Ts of the LED package was around 80℃; moreover, the Ts of the diffuser was 70℃ and that of the reflector was 75℃.

The color shift of the luminaire was investigated using an approach based on the view-factor method (refer to [31] for details). To illustrate this approach, a schematic of the light paths in the investigated downlight is shown in Figure 3. Each exchange of light in the light paths is considered to contribute to the color shift. The matrix equation, i.e., Equation (4) shown below, can be used to describe the exchanges.

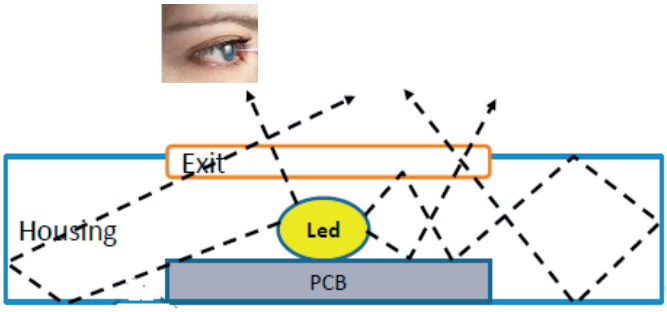


Figure 3 *Schematic of the light paths observed in the investigated downlight*

$$\begin{pmatrix} \Phi_{itoLEDs} \\ \Phi_{itopcb} \\ \Phi_{itohousing} \\ \Phi_{itoexit} \end{pmatrix} = \begin{pmatrix} F_{L-L} & F_{P-L} & F_{h-L} & F_{e-L} \\ F_{L-P} & F_{P-L} & F_{h-p} & F_{e-p} \\ F_{L-h} & F_{P-h} & F_{h-h} & F_{e-h} \\ F_{L-e} & F_{P-e} & F_{h-e} & F_{e-e} \end{pmatrix} \begin{pmatrix} \Phi_{ifromLEDs} \\ \Phi_{ifrompcb} \\ \Phi_{ifromhousing} \\ \Phi_{ifromexit} \end{pmatrix} \quad (4)$$

Where Φ represents light flux, i denote the index of the exchange, and F is the view factor. By conservation of light, the sum of each matrix column is equal to 1. The Light Tools software can be used to extract F and Φ. The physics of reflection can be described by Equation (5).

$$\begin{pmatrix} \Phi_{i+1\,fromLEDs} \\ \Phi_{i+1\,frompcb} \\ \Phi_{i+1\,fromhousing} \\ \Phi_{i+1\,fromexit} \end{pmatrix} = \begin{pmatrix} R_L & & & \\ & R_p & & \\ & & R_h & \\ & & & R_e \end{pmatrix} \begin{pmatrix} \Phi_{itoLEDs} \\ \Phi_{itopcb} \\ \Phi_{itohousing} \\ \Phi_{itoexit} \end{pmatrix} \quad (5)$$

The reflectance properties of reflective materials vary with the wavelength

of the incident light. Therefore, each reflection changes the flux ratio of blue light to yellow light and is therefore accompanied by a small color shift ($\Delta u_i'$, $\Delta v_i'$) depending on the reflective part, i.e., the LED (L), the pcb (p), the housing (h), or the exit (e).

The light transmitted from the exit, $\Phi_{fromexit}$, can be described by Equation (6).

$$\Phi_{fromexit} = (1 - R_e - A)\Phi_{toexit} \tag{6}$$

It is accompanied by a small color shift due the ratio between the blue light and yellow light transmissions at the exit. Note that Φ_{toexit}, which is the light incident on the exit, can be calculated via Equation (7).

$$\begin{aligned} \Phi_{toexit} &= \sum_i \Phi_{itoexit} \\ &= F_{L-e}\sum_i \Phi_{ifromLEDs} + F_{h-e}\sum_i \Phi_{ifromhousing} + F_{p-e}\sum_i \Phi_{ifrompcb} + F_{e-e}\sum_i \Phi_{ifromexit} \end{aligned} \tag{7}$$

The color shift induced by reflection, ($\Delta u'$, $\Delta v'$), of the light as it travels through the exit can be estimated by Equations (8-9).

$$\Delta u_{ref}' = w_{LED} \bullet \Delta u_L' + w_{pcb} \bullet \Delta u_P' + w_{housing} \bullet \Delta u_h' + w_{housing} \bullet \Delta u_e' , \tag{8}$$

$$\Delta v_{ref}' = w_{LED} \bullet \Delta v_L' + w_{pcb} \bullet \Delta v_P' + w_{housing} \bullet \Delta v_h' + w_{housing} \bullet \Delta v_e' , \tag{9}$$

Where w_{LED}, w_{pcb}, $w_{housing}$, and w_{exit} can be described as follows:

$$w_{LED} = \frac{F_{L-e}\sum_i \Phi_{ifromLEDs}}{\Phi_{toextt}}$$

$$w_{pcb} = \frac{F_{p-e}\sum_i \Phi_{ifrompcb}}{\Phi_{toexit}}$$

$$w_{housing} = \frac{F_{h-e}\sum_i \Phi_{ifromhousing}}{\Phi_{toexit}}$$

$$w_{exit} = \frac{r\sum_{i-1}\Phi_{ifromexit}}{\Phi_{toexit}}.$$

Note that ΔuL', Δup', Δuh', and Δue' denote the color shifts associated with the flux in parts L, p, h, and e, respectively; r is the ratio of the flux returned to the exit after one or more reflections of light. All of the light reflections can be simulated by a Monte Carlo simulation using the Light Tools software.

Note that $\Delta u_{tran'}$ and $\Delta v_{tran'}$ are used to express the color shift associated with the exit diffuser, which can be also characterized using a Monte Carlo simulation. The color shift induced by degradation of the LED package is represented by ($\Delta u_{LED}'$, $\Delta v_{LED}'$).

The color shift of the luminaire ($\Delta u_{Lum}'$, $\Delta v_{Lum}'$), is due to the color shift of the LED packages, reflection of the downlight, and transmission through the diffuser. Equations (10-12) describe the magnitude of the color shift in the luminaire.

$$\Delta u_{Lum}' = \Delta u_{LED}' + \Delta u_{ref}' + \Delta u_{tran}', \tag{10}$$

$$\Delta v_{Lum}' = \Delta v_{LED}' + \Delta v_{ref}' + \Delta v_{tran}', \tag{11}$$

$$\Delta u'v'_{Lum} = \sqrt{(\Delta u_{Lum}')^2 + (\Delta v_{Lum}')^2}. \tag{12}$$

Since the LED packages, the reflectance of materials in the reflector, and the transmittance of the exit diffuser degrade with an increase in the aging temperature, the color shift of luminaires depends on their respective degradation mechanisms. For each individual component, the acceleration model can be fit with the Arrhenius equation.

To perform simulations for view factor extraction, the initial condition is shown in Equation (13).

$$\begin{pmatrix} \Phi_{0\,fromLEDs} \\ \Phi_{0\,frompcb} \\ \Phi_{0\,fromhousing} \\ \Phi_{0\,fromexit} \end{pmatrix} = \begin{pmatrix} \Phi_{LEDs} \\ 0 \\ 0 \\ 0 \end{pmatrix}. \tag{13}$$

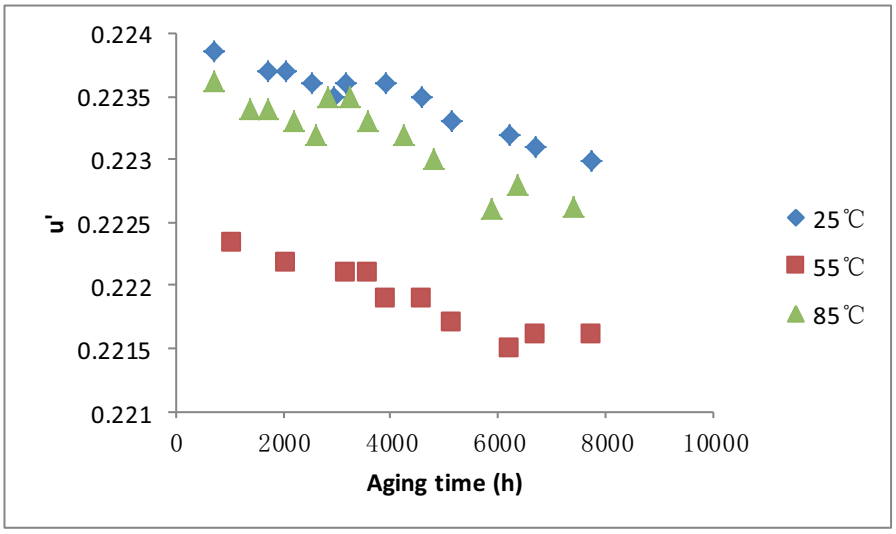
The following assumptions were made: the LED is a top-emitting LED, light exits from the LEDs, and there is no bouncing back and forth between LEDs.

7.3 Results and Discussions

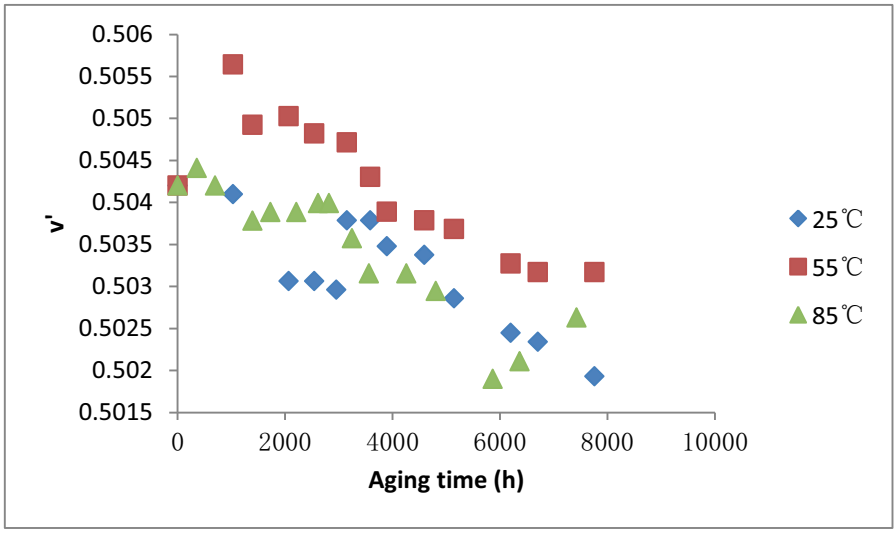
7.3.1 LED Package

7.3.1.1 LED Package Color Shift

Chromaticity coordinate data, u' and v', was obtained during aging for different aging conditions, as shown in Figure 4 (a) and (b), respectively. Results show that both u' and v' decrease with aging time, as expected.



(a) u'



(b) v'

Figure 4 *Color shift with different aging conditions*

3.1.2 Color shift fitting and prediction due to LED package degradation

The exponential model in Equation (14) and the Arrhenius equation (15) are used to fit the degradation of u' and v'.

$$u'(t) = \beta e^{-\alpha t} \tag{14}$$

$$\alpha(T) = A \exp(-Ea / k_b T) \tag{15}$$

Tables 1 and 2 show the parameters calculated based on the data in Figure 4, which may be used for color-shift prediction.

Table 1 Parameters for u' (LED Package)

Ts	62℃	75℃	85℃
α	5.36E-07	5.78E-07	6.67E-07
β	1.0029	0.9963	1.0019
Ea	0.103eV		
A	1.87E-05		

Table 2 Parameters for v' (LED Package)

Ts	62℃	75℃	85℃
α	5.22E-07	5.68E-07	6.30E-07
β	1.0000	1.0021	1.0005
Ea	0.086eV		
A	1.03E-05		

When the ambient temperature, Ta, of the luminaire is 25℃, the Ts inside the LED package is 80℃, calculated using α=6.36E-07 and 6.06E-07 for u' and v', respectively. The color shift of the LED package mounted in the luminaire can be extrapolated and expressed by Equations (16-17), as shown in Figure 5 up to 30000 h of aging.

$$\Delta u_{LED}'(t) = u_{LED}'(0)(e^{-6.36E-07t} - 1) \tag{16}$$

$$\Delta v_{LED}'(t) = v_{LED}'(0)(e^{-6.06E-07t} - 1) \tag{17}$$

$u_{LED}'(0)$ and $v_{LED}'(0)$ represent the original color of the LED package without aging, averaging 0.2233 and 0.5042 respectively.

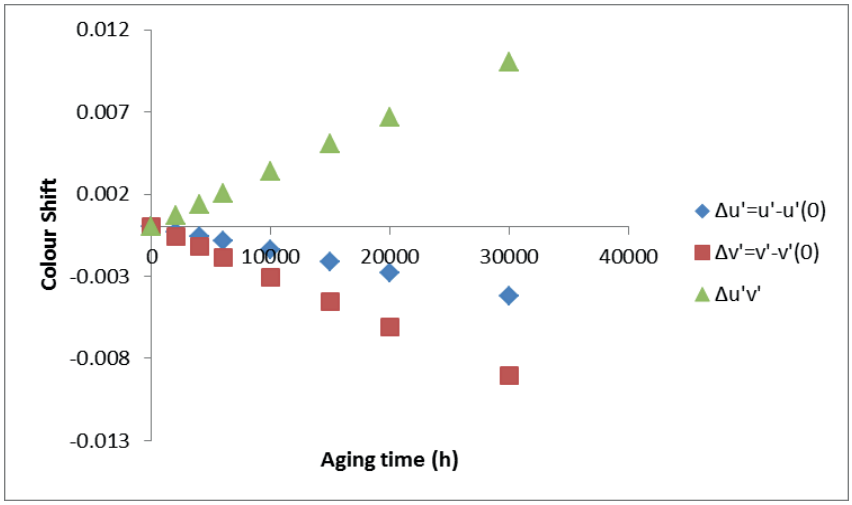
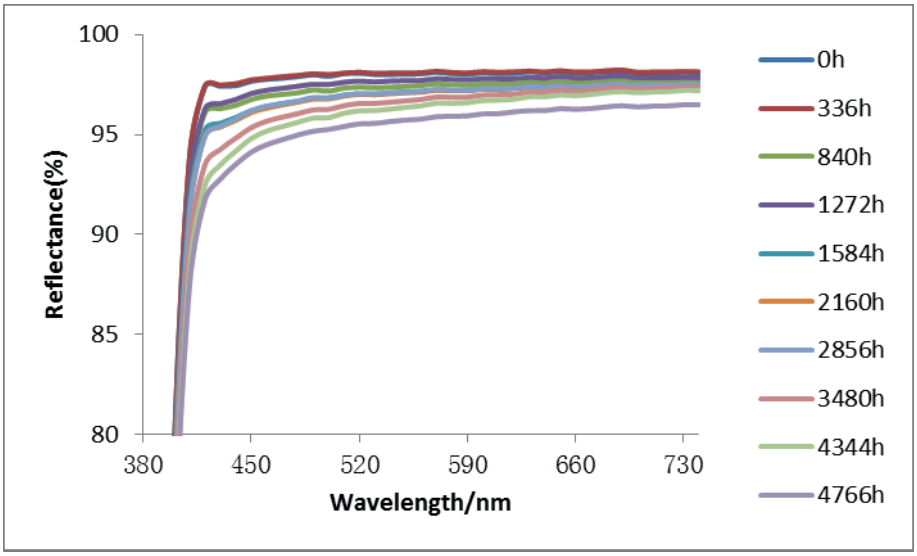


Figure 5 *Color shift extrapolation for the LED package degradation in the luminaire (Ta=25℃)*

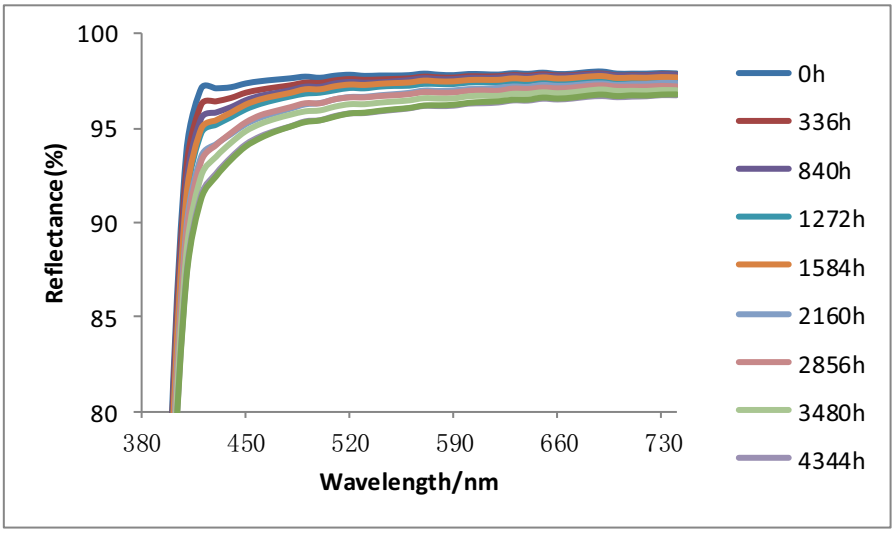
7.3.2 Reflector (MCPOLYCA)

7.3.2.1 Reflectance Degradation

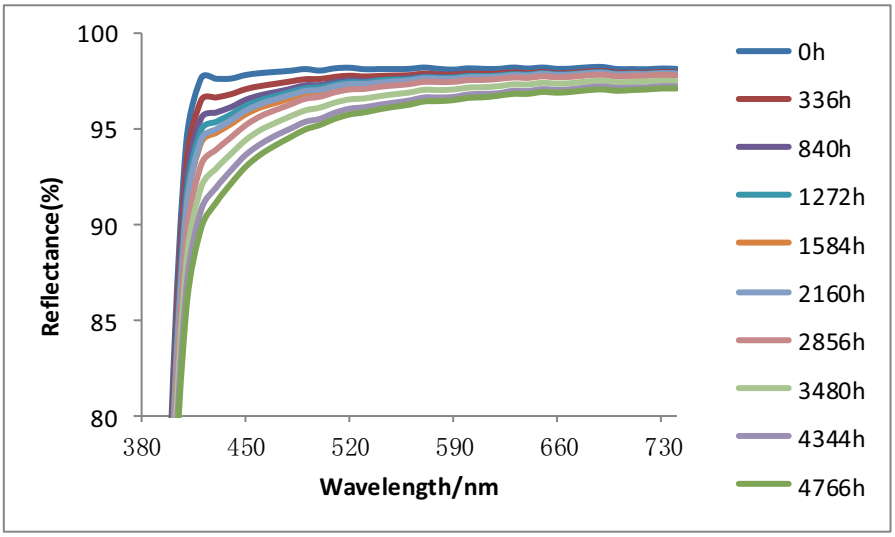
The light reflection was measured and shown in Figure 6 for the MCPOLYCA samples aged under different conditions. Results show that the reflectance decreases with aging time for all samples; the reflectance of blue light (about 450 nm) decreases significantly more than that of yellow light (about 585 nm). This differential reflectance between blue and yellow wavelengths results in a color shift associated with the reflection.



(a) 55℃



(b) 85℃



(c) 100℃

Figure 6 *MCPOLYCA degradation at different temperatures*

7.3.2.2 Degradation of the reflectance ratio for wavelengths between 450 and 585 nm

The degradation of the reflectance ratio for wavelengths between 450 and 585 nm is demonstrated in Figure 7 with different aging temperatures. Based on the results, the reflectance ratio in the wavelength of 450 to 585 nm decreases with aging time, and this degradation intensifies at higher temperatures.

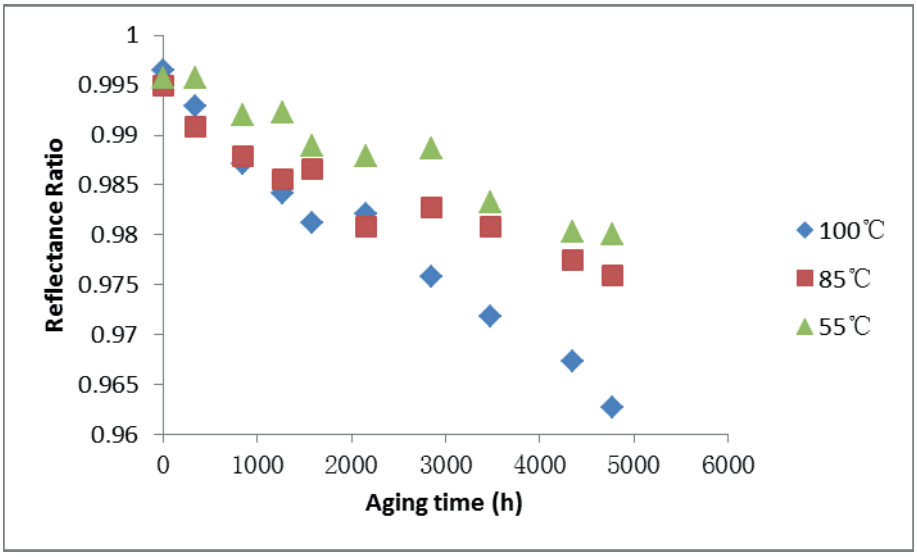
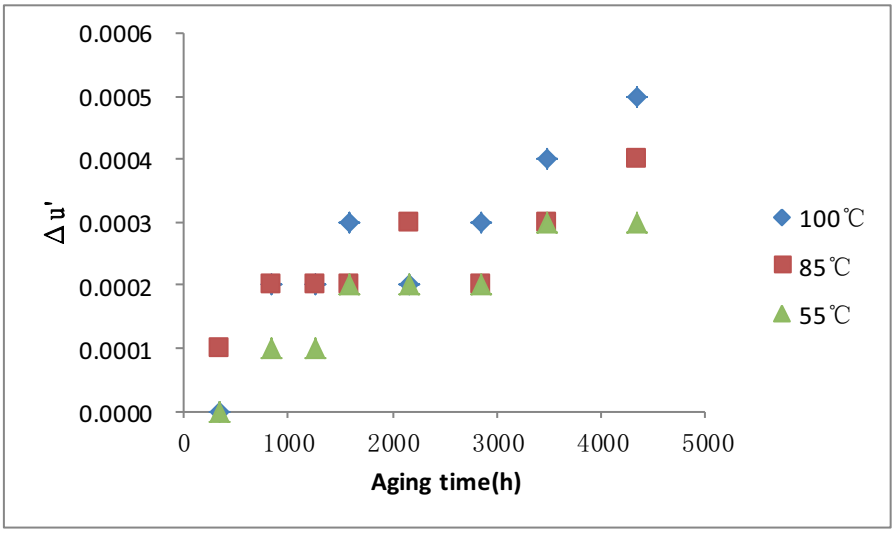


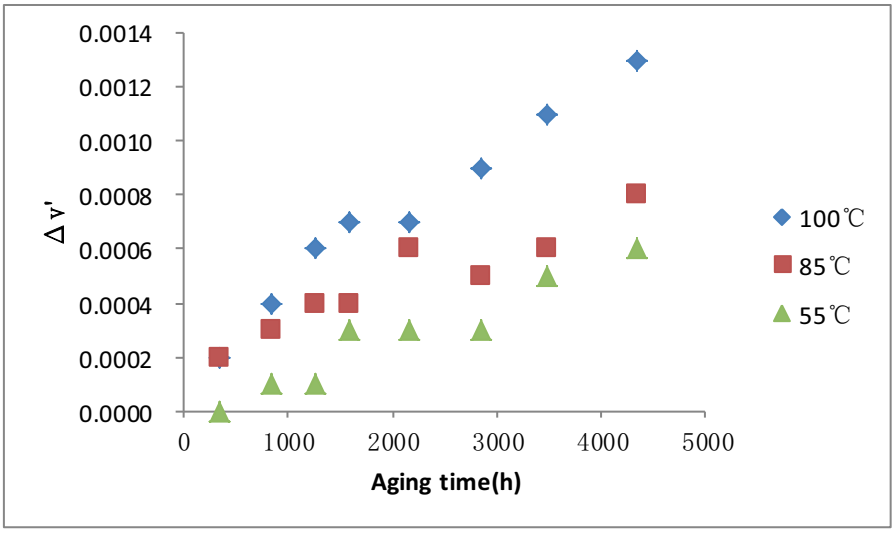
Figure 7 *Degradation of the reflectance ratio for wavelengths between 450 and 585 nm with different aging temperatures*

7.3.2.3 Color shift at different aging conditions

The color shift of the reflected light in terms of Δu' and Δv' was simulated by Monte Carlo modeling; the results are shown in Figure 8 (a) and (b), respectively. Both Δu' and Δv' increase with aging time, and the temperature induces an obvious acceleration effect.



(a) Δu'



(b) Δv'

Figure 8 *Color shift (in terms of Δu' and Δv') due to the degradation of reflectance with different aging conditions*

7.3.2.4 Fitting and prediction of the color shift due to MCPOLYCA degradation

Assuming the degradation follows the exponential model and the reaction rate has an Arrhenius relationship with temperature, the parameters for predicting the color shift in terms of u' and v' can be calculated based on the measured data, and are shown in Tables 3 and 4, respectively.

Table 3 Parameters for u' (reflection degradation)

	100℃	85℃	55℃
α	5.02E-07	4.18E-07	2.96E-07
β	1.0001	1	1.0001
Ea	0.123 eV		
A	2.26E-05		

Table 4 Parameters for v' (reflection degradation)

	100℃	85℃	55℃
α	5.60E-07	3.76E-07	2.68E-07
β	1.0003	1.0000	1.0000
Ea	0.173 eV		
A	1.23E-04		

When the ambient temperature, Ta, of the luminaire is 25℃, the average temperature of the reflectors (including the bottom reflector covering the PCB and the reflector attached to the side wall, both of which are made from MCPOLYCA) is around 75℃, which is calculated using α=3.798E-07 and 3.802E-07 for Δu' and Δv' respectively. The color shift due to MCPOLYCA degradation shown in Figure 9 (up to 30000h), can be extrapolated and expressed by Equations (18-19).

$$\Delta u_{MCP}'(t) = u_{MCP}'(0)(e^{3.798E-07t} - 1) \quad (18)$$

$$\Delta v_{MCP}'(t) = v_{MCP}'(0)(e^{3.802E-07t} - 1) \quad (19)$$

Where $u_{MCP}'(0)$ and $v_{MCP}'(0)$ are the color values of the light after being reflected by the unaged MCPOLYCA material, and are assumed to be 0.2233 and 0.5044, respectively, per the Monte Carlo simulation.

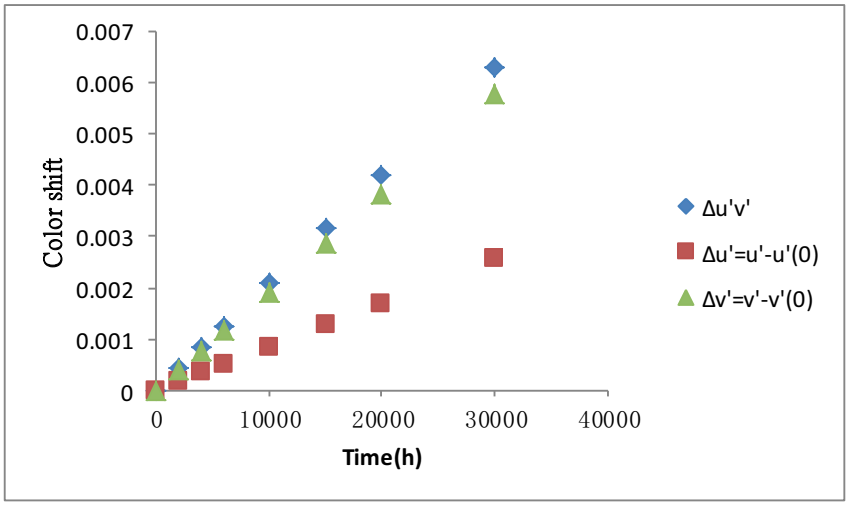


Figure 9 *Extrapolation of color shift during aging at 75℃ (where Ta=25℃) due to degradation in the reflector*

It is important to point out that the magnitude of the color shift extrapolated in Equations (18-19) denotes the color shift of the light after reflection by the MCPOLYCA reflectors. Part of the light emitted from the LED packages doesn't undergo reflection and therefore will not undergo this color shift. The effect of reflection on the luminaire color shift is described by Equations (8-9).

7.3.3 Diffuser

Discoloration was observed on the surfaces of the samples subjected to aging. The discoloration of the samples after 4000 h of aging is shown in Figure 10. The discoloration is most significant following aging at 115℃.

7.3.3.1 Diffuser degradation

Light transmittance through the diffuser was measured during aging, as shown in Figure 11. Based on the data, the degradation of the diffuser increases with time and temperature.

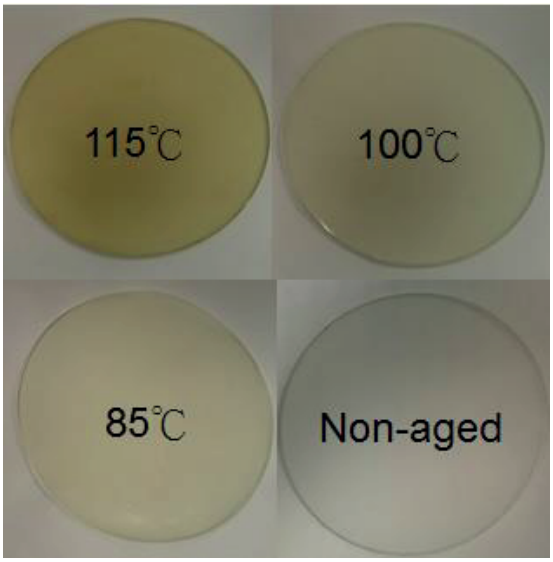
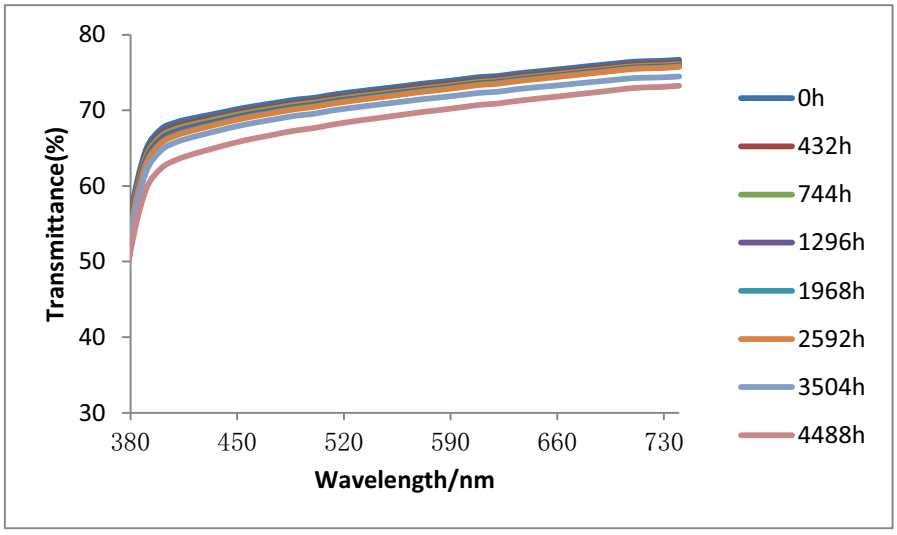
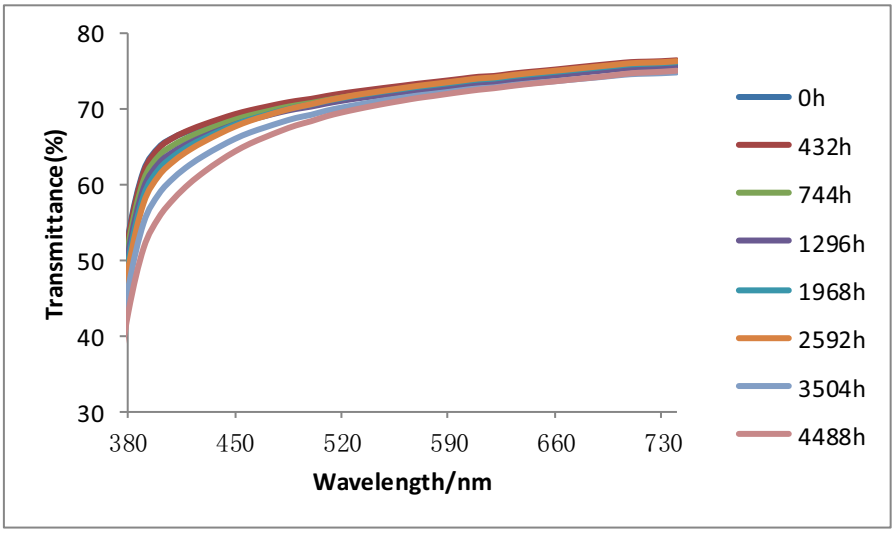


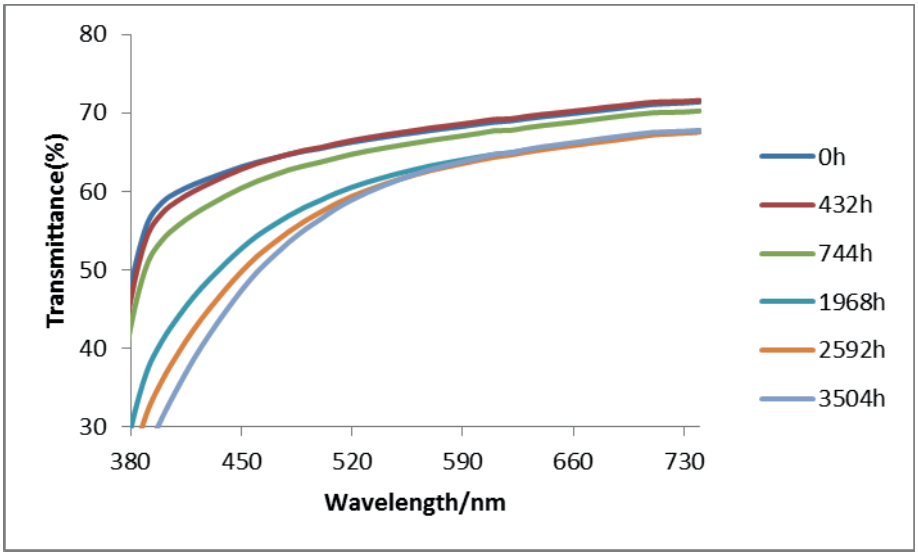
Figure 10 *Discoloration of samples subjected to different aging conditions for around 4000 h*



(a) 85℃



(b) 100℃



(c) 115℃

Figure 11 *Diffuser degradation at different temperatures*

7.3.3.2 Degradation of transmittance ratio for wavelengths between 450 and 585 nm

The degradation of the transmittance ratio for wavelengths between 450 and 585 nm under different aging temperatures is shown in Figure 12. As with the degradation of the reflector, the ratio of transmittance for wavelengths between 450 and 585 nm decreases with aging time, and is enhanced at higher temperatures.

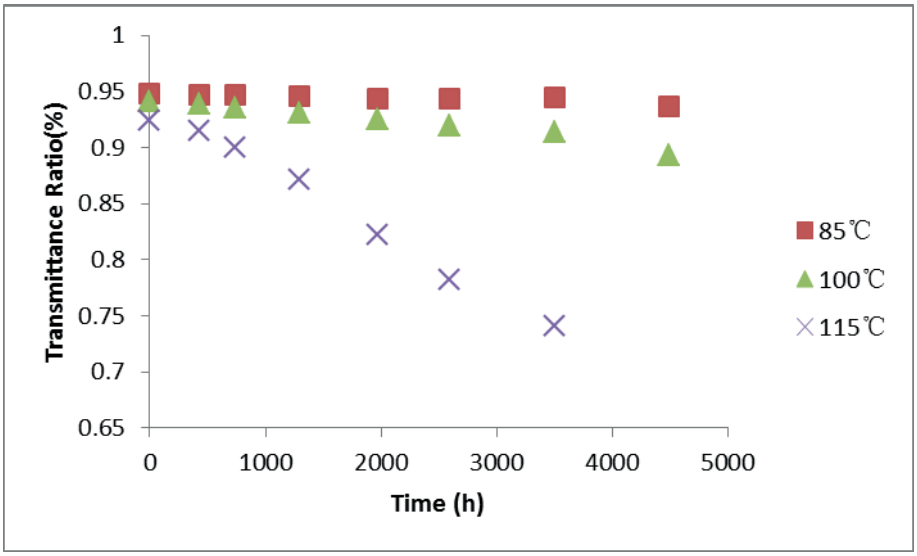
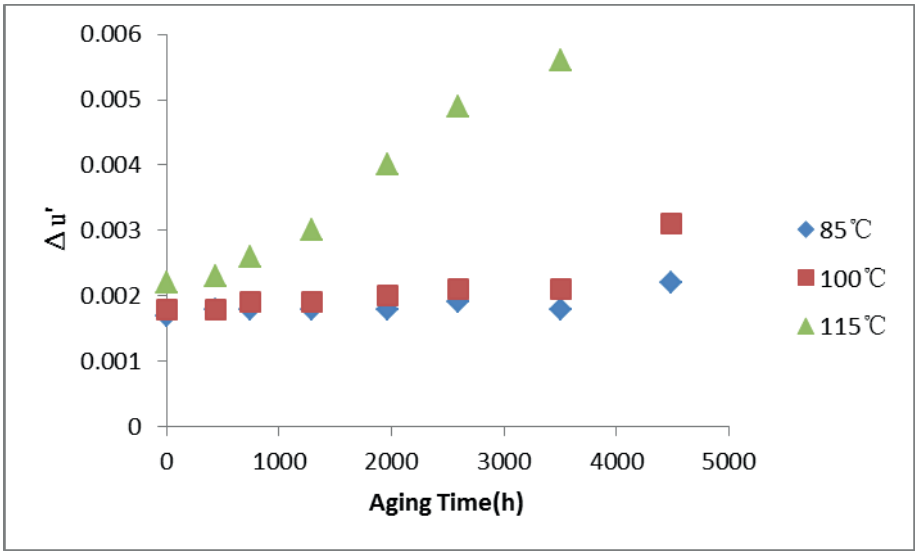


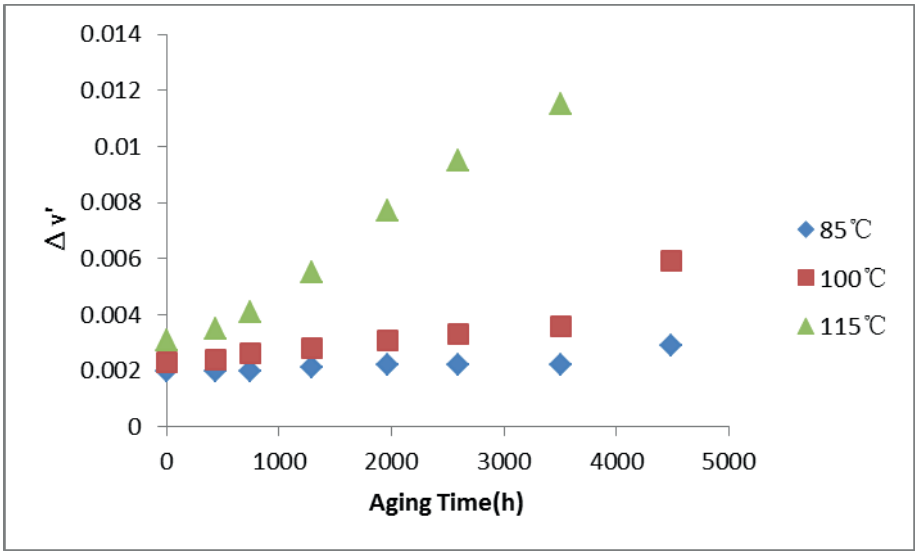
Figure 12 *Degradation of transmittance ratio for wavelengths between 450 and 585 nm under different aging temperatures*

7.3.3.3 Color shift due to different aging conditions

The color shift due to the diffuser degradation in terms of Δu' and Δv' were also simulated by the Monte Carlo modeling; the results are shown in Figure 13 (a) and (b) respectively. As observed with the degradation of the reflector, both Δu' and Δv' increase with aging time, and the temperature caused a notable acceleration of this effect.



(a) Δu'



(b) Δv'

Figure 13 *Color shift (Δu' and Δv') under different aging conditions*

7.3.4 Fitting and prediction of color shift due to transmittance degradation

Table 5 Parameters for u' (diffuser degradation)

	85°C	100°C	115°C
α	3.36E-07	1.01E-06	4.66E-04
β	0.9636	0.9500	1.0118
Ea	0.904 eV		
A	2.16E+06		

Table 6 Parameters for v' (diffuser degradation)

	85°C	100°C	115°C
α	3.18E-07	1.32E-06	5.02E-06
β	0.9600	0.9565	1.0097
Ea	0.927eV		
A	5.48E+06		

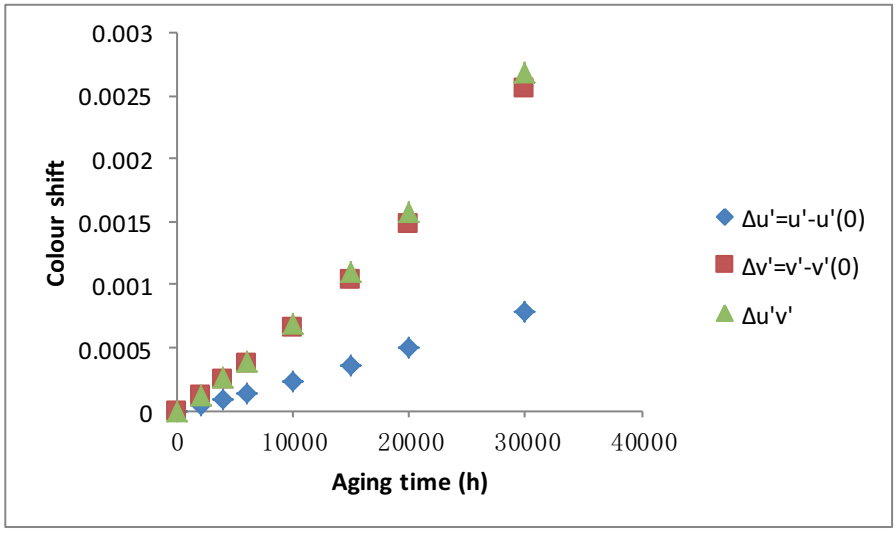


Figure 14 *Extrapolation of the color shift due to aging at 70 ℃ (with Ta=25 ℃) due to transmittance degradation*

As with the reflector, assuming that the degradation follows an exponential model and the reaction rate has an Arrhenius relationship with temperature, parameters for color shift can be calculated based on the recorded data, and are shown in Tables 5 and 6, respectively.

When the ambient temperature, Ta, of the luminaire is 25℃, the temperature inside the diffuser is 70℃, as calculated assuming α=1.12E-07 and 1.32E-07 for u' and v', respectively. The color change due to degradation of the diffuser transmittance can be extrapolated as shown in Figure 14 (up to 30000 h) and expressed by Equations (17-18).

$$\Delta u_{tran}'(t) = u_{tran}'(0)(e^{1.12E-07t} - 1) \tag{20}$$

$$\Delta v_{tran}'(t) = v_{tran}'(0)(e^{1.32E-07t} - 1) \tag{21}$$

where $u_{tran}'(0)$ and $v_{tran}'(0)$ are the color values of the transmitted light that passed through the unaged diffuser, and are assumed to be 0.2251 and 0.5065, respectively, according to the Monte Carlo simulation.

7.3.4 Luminaire Color Shift Prediction

The color shift of the luminaire is due to a combination of the effects of the degradation of the LED packages, reflectors, and diffuser. The color shift of the luminaire which is contributed by the LED package can be predicted using Equations (16-17), and that of the diffuser can be predicted by Equations (20-21). However, since only a portion of the light in the downlight undergoes reflection, the contribution of the reflector to the color shift of the luminaire cannot be predicted by Equations (18-19) alone. Instead, Equations (8-9) can be used to calculate the color shift caused by reflection in the downlight. To simplify this calculation, the light flux bouncing from LED packages and the color shift induced by the small amount of reflection by the diffuser are disregarded. The view factors (F) were extracted via the Monte Carlo modelling and are shown in the table 7. Based on this information, the corresponding prefactors, w_{pcb} and $w_{housing}$, in Equations (8-9) can be calculated accordingly as 0.128 and 0.249, respectively. Since the PCB and the side-wall of the luminaire are covered by the reflective material, MCPOLYCA, the color shift due to reflection can be simplified as shown in Equations (22-23).

$$\Delta u_{ref}'(t) = (w_{pcb} + w_{housing})\Delta u_{MCP}'(t) = 0.377 u_{MCP}'(0)(e^{3.798E-07t} - 1) \tag{22}$$

$$\Delta v_{ref}'(t) = (w_{pcb} + w_{housing})\Delta v_{MCP}'(t) = 0.377 v_{MCP}'(0)(e^{3.802E-07t} - 1) \tag{23}$$

Table 7 View factors for color-shift investigation

		From			
		LEDs	PCB	Housing	Exit
To	LEDs	0.000	0.000	0.000	0.000
	PCB	0.000	0.000	0.245	0.533
	Housing	0.214	0.214	0.000	0.467
	Exit	0.786	0.786	0.755	0.000

The luminaire color shift for room temperature applications (Ta=25°C) can be predicted by Equations (24-26),

$$\Delta u_{Lum}'(t) = \Delta u_{LED}'(t) + \Delta u_{ref}'(t) + \Delta u_{tran}'(t) \quad (24)$$

$$\Delta v_{Lum}'(t) = \Delta v_{LED}'(t) + \Delta v_{ref}'(t) + \Delta v_{tran}'(t) \quad (25)$$

$$\Delta u'v'_{Lum}(t) = \sqrt{(\Delta u_{Lum}'(t))^2 + (\Delta v_{Lum}'(t))^2} \quad (26)$$

Where the contributions from the LED packages, $\Delta u_{LED}'(t)$ and $\Delta v_{LED}'(t)$, the reflector reflectance, $\Delta u_{ref}'(t)$ and $\Delta v_{ref}'(t)$, and the diffuser transmittance, $\Delta u_{tran}'(t)$ and $\Delta v_{tran}'(t)$, can be determined by Equations (16-17), (22-23), and (20, 21) respectively. The prediction of the color shift over the lifetime of the down light is shown in Figure 15 (up to 30000h).

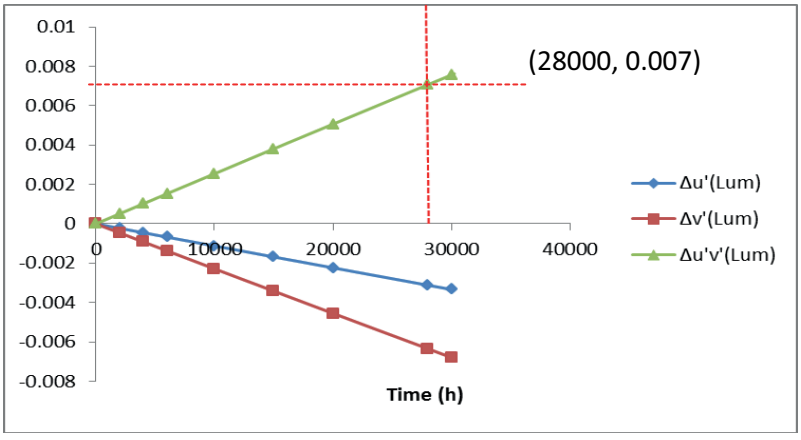
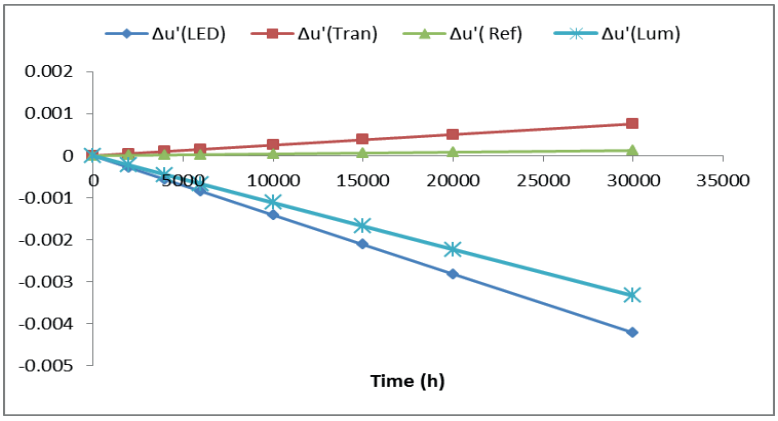
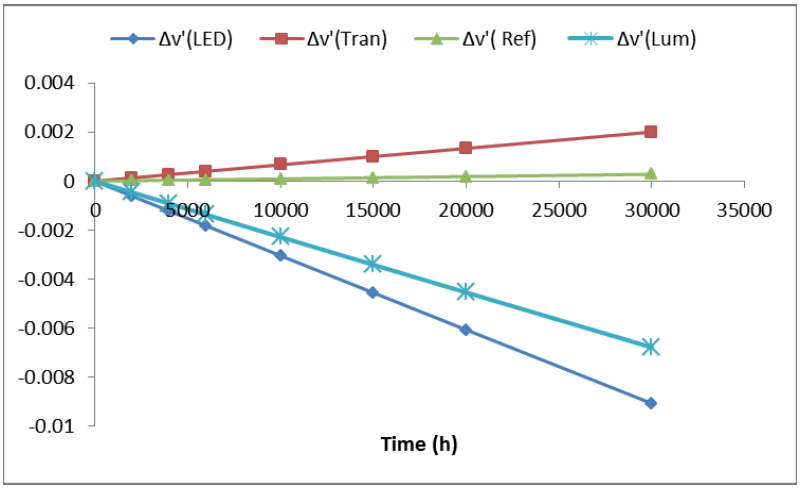


Figure 15 *Color shift prediction on the luminaire investigated*

The results show that both Δu' and Δv' of the luminaire are negative and the magnitude of the luminaire color shift, Δu'v', increases with aging time up to 0.007 at 28000 h. This indicates a blue shift in the luminaire color. The color shifts caused by each of the individual components in terms of Δu' and Δv' are shown in Figure 16 (a) and (b), respectively.



(a) Δu'



(b) Δv'

Figure 16 *Contributions of the components to the color shift of the luminaire*

The color shift contributed by each individual component varies greatly in quantity and direction. The color shifts contributed by the reflector and the diffuser both have positive Δu' and Δv' values, indicating a yellow shift, and will increase with aging time. The contribution from the diffuser degradation is more significant than the contribution by the degradation of the MCPOLYA reflectors. By contrast, the color shift due to the LED package degradation has negative Δu' and Δv' values, indicating a blue shift in the color, and the absolute value of this shift increases with aging time. The overall color shift of the LED luminaire is toward blue, indicating that the LED package dominates the color shift. It is interesting to note that the color shifts caused by LED packages counteracted that of the diffuser and reflectors in terms of both u' and v'. Thus, the overall color shift of the luminaire is less than that caused by the LED packages alone.

7.4 Conclusions

The acceleration and prediction method used in this paper can be conveniently leveraged to investigate the color shift of the luminaire due to of each individual part, and to predict the overall color shift. Based on the results of this investigation, it can be concluded that:

1) The color shift induced by aging of the LED packages is quite different from that of diffuser and reflector in magnitude and direction. The color LED packages cause a blue shift and the reflector and diffuser cause a yellow shift in the color.

2) The overall color shift of the LED-based luminaire is toward blue, as the LED packages dominate the color shift of the luminaire investigated here.

3) The color shift caused by the LED packages and that by the diffusers and reflectors counteract each other, so the total color shift of the luminaire is lower than the color shift of LED packages alone.

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Chapter 8

Design Rules for LED Based Luminaire Color Maintenance

Conclusions are given on the basis of Chapter 2 ~7. According to investigation results and conclusions in this thesis, some design rules for color maintenance of LED luminaires are given. Recommendations for the future studies include further study on the degradation kinetics of LED Packages, root cause study for color variation and diffuser's reflectivity degradation effect on the luminaire color shift prediction

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8.1 Design Rules-1

Color shift mechanisms are investigated for each individual part in the LED based luminaire: diffuser, reflector and LED packages. In order to quantify the color shift caused by each individual part, a novel approach is proposed and used in a downlight for further investigation. Finally, an acceleration method and accompanying prediction for luminaire level color shift is proposed and investigated using an LED based down light. Conclusions could be drawn as below:

Diffuser materials (BPA-PC and PMMA) are experimentally investigated on the color shift effects during aging, and color shift mechanisms are also studied. Results revealed:

- Inconsistent degradation of wavelength–dependent transmittance induces the decrease of the blue/yellow light intensity ratio and thus gives rise to the color shift toward the yellow field, which is the color shift mechanism of BPA-PC;
- Even for the non-aged BPA-PC, the transmittance varies with wavelength in the visible light field due to the chemistry of the material, which caused the change of intensify ratio of blue light to yellow light in the SPD, leading to color change in perception;
- Oxidation plays a key role in the degradation of transmittance at around the peak wavelength of the blue light field, which is in correlation with the discoloration of thermally-aged BPA-PC materials.

By contrast, for the PMMA specimen aged up to 3000 hours, oxidation was neither occurred at 85deg.C, nor with additional exposure to blue light, nor even with additional humidity of 85%RH. From further investigation on the PMMA diffuser, below conclusions could be drawn:

- Discoloration was not observed for any sample subjected to aging of 85℃ for 5000 hours, or with additional blue light irradiation for 5000 hours, or with additional humidity of 85%RH for 5000 hours, or even with aging of 100℃ for 3000 hours.

- The specimen subjected to aging of 150℃ for 360 hours has a surface discoloration and has a significant wavelength dependent degradation in the transmission spectrum caused by oxidation. The specimen with aging of 100℃ for 3000 hours has a less oxidation, although no significant transmission spectrum reduction was observed.
- Using such aged specimen as a diffuser mounted on a LED-based luminaire, the radiant flux peak intensity in the blue light area has a more severe reduction than that in the yellow light area, which results in a reduction of the radiant flux intensity ratio of blue light to yellow light and hence induces the color shift to yellow.

Failure modes and degradation mechanisms for *Microcellular PET reflective materials* are investigated under different conditions. Results show that:

- A humidity test at 85 ºC & 85%RH for 4000hours (or even shorter) can lead to hydrolytic degradation, which causes both the decrease of reflectance varying with wavelength and a severe embrittlement;
- Oxidative degradation occurred at 85 ºC for 4000hours can cause a slight reflectance spectrum change, while the additional blue light exposure has little impact;
- Color shift induced by thermal aging at 85 ºC for 4000hours is 0.0001 and lumen efficiency decreased by 1.57%. When Microcellular PET crumbles due to the embrittlement during humidity test, the color shift increases to 0.0004 and the lumen efficiency is reduced by 4.47%.

Color shifts of widely used *Mid-power white light LED packages* under different aging conditions are investigated. Temperature stress, humidity stress and current stress are experimentally designed and performed to accelerate the color shift of mid-power LED packages and color shift mechanisms have been discussed based on the color shift results obtained from measurements. Conlusions could be drawn: Exponential fitting demonstrates a good exponential relationship between color shift (Δu', Δv') and aging time almost for all the aging conditions. We can extrapolate the color shift Δu' and Δv' based on the fitted regression equations and then make the prediction for the total color shift Δu'v'.

Current stress can induce a different failure mode. Peak intensity reduction analysis reveals that the current stress accelerates the degradation of LED die. Humidity test induced a substantial color shift both in u'and v'. Peak intensity comparison analysis reveals that not only the degradation of LED die, but also degradation of the phosphor layer contributes to such high color shift in the humidity test.

In order to *quantify the color shift caused by each individual component* in the LED based luminaire, a novel approach is proposed for color shift investigation on LED-based Luminaires. Using this approach, color shift contribution of each individual component (diffuser, reflector, housing and LED package) could be calculated besides luminaire level color shift. This approach is based on a view factor method. Conclusions could be drawn after an investigation on an LED downlight before and after aging at 85℃ for around 4000 hours. The approach proposed in this paper can be easily used to extract the color shift contribution of each individual component for LED luminaires. According to the results from the simulation and calculation method, after aging of 4000h, the downlight has an overall color shift (Δu'v') of ~0.002, even less than the color shift of LED packages, which is around 0.0025. This is because the two major contributors LED package and diffuser have an opposite contribution in the color shift component of Δv'. The component Δv' of LED package has a decrease of 0.0026, compared to the increase of around 0.0017 caused by diffuser transmission change during aging. Color shift induced by aging of LED packages is quite different from that of diffuser. LED packages' relative flux degradation in SPD occurred obviously in the yellow light area, which induces the color shift to blue. By contrast, the color will shift to yellow for aging of the diffuser since there is much more degradation of flux in the blue light area.

LED based luminaire color shift acceleration method and prediction approach are illustrated and proposed. Prediction results were also presented. Conclusions could be drawn as:

- Color shift induced by aging of LED packages is quite different from that of diffuser and reflector, LED packages' relative flux degradation in SPD occurred obviously in the yellow light area, which induces the color shift to blue.

- By contrast, the color will shift to yellow for aging of the diffuser and reflector since there is much more degradation of flux in the blue light area. Since the color shift caused by LED packages and by diffusers (and reflectors) cancelled out each other, the total color shift of the luminaire is even less than the color shift of LED packages.
- The method used in this thesis could be used for the LED-based luminaire color shift prediction.

8.2 Design Rules-2

According to investigation results and conclusions in this thesis, color shift mode induced by aging of LED packages is quite different from that of diffuser and reflector, LED packages' relative flux degradation in SPD occurred obviously in the yellow light area, which induces the color shift to blue. By contrast, the color will shift to yellow for aging of the diffuser and reflector since there is much more degradation of flux in the blue light area. Since the color shift caused by LED packages and by diffusers (and reflectors) cancelled out each other, the total color shift of the luminaire is even less than the color shift of LED packages. In addition, current stress can also induce a different failure mode, v' may be increased due to high current, compared to a decrease of v' in the normal current loading. Based on our findings, design rules for indoor illumination applications are drafted as below:

- When we use the PMMA as the diffuser and PET or MCPOLYCA as the reflector, and same type of LED packages as the light source, if the color maintenance of LED package can meet the requirement of DOE Energy Star Program, the color maintenance of the luminaire designed can also meet that requirement under normal operation conditions.
- If the application is over current loading, there will be an increase in v' for the LED package instead of decrease in normal current loading, since both reflector and diffuser also contribute positive shift in v', the color shift of luminaire in v' will be increased much and induce a color maintenance problem. In these cases, the lifetime of the product is not limited by lumen maintenance, rather by color shift.

- If Target lifetime (here defined as the color shift is below 0.007) of the LED based luminaire is 20000h or 30000h, we can get the allowed maximum Ts when the minimum temperatures of diffuser and reflctor are given, as shown in Table 1.

Table 1 Target Lifetime Vs. Allowed Ts of LED Package

Target Lifetime(h)	20000				30000	
Allowed Max. Ts(℃)	92	98	113	148	73	110
Ttran (℃)	50	60	70	80	70	80
Tref (℃)	55	65	75	85	73	85

8.3 Recommendations

As mentioned, there is substantial need for accelerated testing to forecast color shift behaviour of LED based luminaires. The IES has initiated a committee with a target toward color shift prediction of LED packages (TM21 alike) and this thesis propose a method to extend component-level data to complete LED lamps and luminaires for color shift prediction. Still many challenges in this area exist, which can be recommended for the future studies.

- *Further Study on the degradation kinetics of LED Packages.* Although the LED package investigated in this thesis can follow the exponential model in color shift and the reaction rate has an Arrhenius relationship with temperature, but this cannot guarantee that other LED packages follow the same kinetics.
- *Root cause study for color variation.* Sample to sample variation, lot to lot variation exist for the LED packages investigated in this thesis, which reveals there should be some inputs which need to be controlled. Root cause analysis will not only be helpful in the color maintenance investigation, but may also give some clues for the color shift prediction to find ways to control it.
- *Diffuser's reflectivity degradation effect on the luminaire color shift prediction.* Although the color shift caused by the reflectivity degradation of diffuser is investigated in Chapter 6, such effect is

neglected in Chapter 7 for color shift acceleration and prediction for the sake of simplicity. Effects could be further investigated.

List of Abbreviations and Symbols

SSL	Solid State Lighting
LED	Light Emitting Diode
CIE	Commission International de l'Eclairage
CSA	China Solid State Lighting Alliance
SPD	Spectral Power Distribution
CCT	Correlated Color Temperature
T	Temperature
t	Time
Tc	Case Temperature
Ts	Soldering Temperature
x	The xyz coordinates
y	The xyz coordinates
u'	The (u', v')chromaticity
v'	The (u', v')chromaticity
Δu'	Chromaticity change in u' coordinates
Δv'	Chromaticity change in v' coordinates
Δu'v'	The magnitude of color shift or change in (u', v')
λ	The wavelength
Φ	The luminous flux
Ea	Activation engergy
α	Reaction rate

Summary

Luminous flux maintenance and color stability are two important factors to evaluate the lighting quality. Where a significant amount of research efforts are performed on the former and even some standards have been generated for lumen decay acceleration, research activities and achievements on the latter are lagging behind. DOE Energy Star Program requires that the change of chromaticity over the minimum lumen maintenance test period (6000 hours) should be within 0.007 on the CIE 1976 (u', v') diagram. The color shift mechanisms for LED lighting are complex due to its comprehensive structure, generally composed of LED die, phosphor, silicone, reflector, diffusers, and so on, all of which may contribute to the color shift during operation. On top of this, each individual component has its own degradation mechanism. Studies on the LED based luminaire level color shift reliability are little publicly available except some based on the statistical (data-driven) method.

There is a substantial need for an accelerated testing method able to predict color shift especially for LED based luminaires. The IES has initiated a committee that will be charged with developing an approved procedure. This work is targeted toward LED packages, for which the exact operating characteristics are more easily controlled. Much like lifetime and reliability, extending component-level data to complete LED lamps and luminaires can be difficult and many challenges exist. A lack of standard procedures for predicting color stability performance has contributed to user uncertainty—potentially limiting adoption and making it more challenging (even impossible) for manufacturers to provide warranty coverage for color shift.

Challenges for LED lamp or luminaire level color shift predictions are: color shift mechanisms and prediction caused by LED packages under different aging conditions, color shift mechanisms and prediction caused by diffusers and reflectors, investigation method for color shift quantification caused by each individual component and an acceleration method or approach for luminaire level color shift.

In order to help overcome those challenges mentioned above, objectives are set up in this thesis as:

1. color shift mechanisms investigation for major diffuser materials: BPA-PC and PMMA
2. color shift mechanisms investigation for major reflective materials: MCPET and MCPOLYCA
3. color shift investigation for mid-power LED packages, investigation method for color shift quantification caused by each individual component in the LED based luminaire
4. acceleration method or approach for luminaire level color shift, and prediction for luminaire level color shift

Color Shift mechanisms for LED Secondary Optical Designs were investigated and comparisons between two widely used diffusers, BPA-PC and PMMA, were made. In this chapter, broadly used commercial diffuser materials (BPA-PC and PMMA) are experimentally investigated towards the color shift effects during aging. Besides this, color shift mechanisms of degradation of transmittance are also studied. Results revealed:

- Inconsistent degradation of wavelength–dependent transmittance induces the decrease of the blue/yellow light intensity ratio and thus gives rise to the color shift toward the yellow field, which is the color shift mechanism of BPA-PC;
- Even for the non-aged BPA-PC, the transmittance varies with wavelength in the visible light field due to the chemistry of the material，which caused the change of intensify ratio of blue light to yellow light in the SPD， leading to color change in perception;
- Oxidation plays a key role in the degradation of transmittance at around the peak wavelength of the blue light field, which is in correlation with the discoloration of thermally-aged BPA-PC materials. By contrast, for the PMMA specimen aged up to 3000 hours, oxidation was neither occurred at 85℃ nor with additional exposure to blue light, nor even with additional humidity of 85%RH.

Color Shift and its mechanism were furtherly investigated on the PMMA diffuser used in LED-based Luminaires, conclusions could be drawn as:

- Discoloration was not observed for any sample subjected to aging of

85℃ for 5000 hours, or with additional blue light irradiation for 5000 hours, or with additional humidity of 85%RH for 5000 hours, or even with aging of 100℃ for 3000 hours.

- The specimen subjected to aging of 150℃for 360 hours has a surface discoloration and has a significant wavelength dependent degradation in the transmission spectrum caused by oxidation. The specimen with aging of 100℃ for 3000 hours has a less oxidation, although no significant transmission spectrum reduction was observed.
- Using such aged specimen as a diffuser mounted on a LED-based luminaire, the radiant flux peak intensity in the blue light area has a more severe reduction than that in the yellow light area, which results in a reduction of the radiant flux intensity ratio of blue light to yellow light and hence induces the color shift to yellow.

Degradation of Microcellular PET reflective materials used in LED-based products was experimentally determined. Both the lumen maintenance and color shift were investigated. Failure modes and degradation mechanisms were also investigated under different conditions. Results shown:

- A humidity test at 85 ºC & 85%RH for 4000hours (or even shorter) can lead to hydrolytic degradation, which causes both the decrease of reflectance varying with wavelength and a severe embrittlement;
- Oxidative degradation occurred at 85 ºC for 4000hours can cause a slight reflectance spectrum change, while the additional blue light exposure has little impact;
- Color shift induced by thermal aging at 85 ºC for 4000hours is 0.0001 and lumen efficiency decreased by 1.57%. When Microcellular PET crumbles due to the embrittlement during humidity test, the color shift increases to 0.0004 and the lumen efficiency is reduced by 4.47%.

Color shifts of widely used Mid-power white light LED packages under different aging conditions were investigated. The type of Mid-power LED package investigated is also widely utilized in the downlight luminaire for indoor illumination applications. Color shift modes and mechanisms caused by different acceleration stresses were investigated and an acceleration and

prediction method was proposed. Temperature stress, humidity stress and current stress were experimentally designed and performed to accelerate the color shift of mid-power LED packages and color shift mechanisms have been discussed based on the color shift results obtained from measurements. Conclusions could be drawn as follows:

- Exponential fitting demonstrates a good exponential relationship between color shift (Δu', Δv') and aging time almost for all the aging conditions. We can extrapolate the color shift Δu' and Δv' based on the fitted regression equations and then make the prediction for the total color shift Δu'v';
- Current stress can induce a different failure mode. Peak intensity reduction analysis reveals that the current stress accelerates the degradation of LED die;
- Humidity test induced a substantial color shift both in u' and v'. Peak intensity comparison analysis reveals that not only the degradation of the LED die, but also degradation of the phosphor layer contributes to such high levels of color shift in the humidity test.

In order to quantify the color shift caused by each individual component in the LED based luminaire, a novel approach was presented for color shift investigation on LED-based Luminaires. Using this approach, color shift contribution of each individual component (diffuser, reflector, housing and LED package) could be calculated besides luminaire level color shift. This approach is based on a view factor method. Conclusions could be drawn after an investigation on an LED downlight before and after aging at 85℃ for around 4000 hours：The approach proposed in this paper can be easily used to extract the color shift contribution of each individual component for LED luminaires. According to the results from the simulation and calculation method, after aging for 4000h, the downlight has a total color shift of ~0.002, even less than the color shift of LED packages, which is around 0.0025. This is because the two major contributors LED package and diffuser have an opposite contribution in the color shift component of Δv'. The component Δv' of LED package has a decrease of 0.0026, compared to the increase of around 0.0017 caused by diffuser transmission change during aging. Color shift induced by aging of LED packages is quite different from that of diffuser. LED packages' relative flux

degradation in SPD occurred obviously in the yellow light area, which induces the color shift to blue. By contrast, the color will shift to yellow for aging of the diffuser since there is much more degradation of flux in the blue light area.

LED based luminaire color shift acceleration method and prediction approach were illustrated and proposed. Prediction results were also presented. Conclusions could be drawn: Color shift induced by aging of LED packages is quite different from that of diffuser and reflector in magnitude and direction. The color will shift toward blue for LED packages. However, the color shift contributed by reflector and diffuser is toward yellow. The overall color shift of luminaire is toward blue, and the LED packages dominate the color shift direction of the LED based luminaire in the investigation. The color shift caused by LED packages and by diffusers (and reflectors) cancelled out each other, the net color shift of the luminaire is even less than the color shift of the LED packages.

In addition, guidelines and design rules were given based on the results and findings in the previous investigation in Chapter 7, which will be helpful in the design or manufacturing of LED based luminaires.

Samenvatting

Lichtsterkte behoud en kleurstabiliteit zijn twee belangrijke factoren om de lichtkwaliteit te evalueren. Er is aanzienlijk aantal onderzoeken op het behoud van lichtsterkte uitgevoerd en zelfs sommige standaarden zijn gegenereerd. Echter onderzoeksactiviteiten op het gebied van kleurstabiliteit zijn vrij schaars. Het DOE Energy Star Programma vereist dat de verandering van de chromaticiteit over de minimale testperiode van 6000uur binnen 0.007 op het CIE 1976 (u', v') diagram liggen. De kleurveranderingsmechanismen voor LED-verlichting zijn complex dankzij de uitgebreide structuur, meestal samengesteld uit LED-chips, fosforen, siliconen, reflectoren, diffusoren, die allemaal kunnen bijdragen aan de kleurverschuiving tijdens gebruik. Bovendien heeft elke individuele component een eigen degradatie mechanisme. Studies omtrent kleurverschuivingen op armatuurniveau zijn weinig openbaar beschikbaar, op een enkele statistische (data-driven) methode na.

Er is een aanzienlijke behoefte aan een versnelde testmethode die kleurverandering kan voorspellen, vooral voor LED-armaturen. De IES heeft een commissie ingesteld die belast zal worden met het ontwikkelen van een goedgekeurde procedure. Dit werk is alleen gericht op LEDs, waarvoor de precieze degradatie kenmerken gemakkelijker worden geregeld. Het uitbreiden naar andere componenten op het niveau van LED-lampen en armaturen kan moeilijk zijn en er bestaan veel uitdagingen. Uitdagingen voor deze kleurverschuivingsvoorspellingen zijn: kleurverschuivingsmechanismen en voorspellingen veroorzaakt door LEDs onder verschillende verouderingsomstandigheden, kleurverschuivingsmechanismen en voorspellingen veroorzaakt door diffusoren en reflectoren, onderzoeksmethode voor kleurverschuivingsquantificering veroorzaakt door elk afzonderlijk bestanddeel en een versnellingsmethode of aanpak voor de kleurverschuiving op armatuurniveau.

Om te helpen de bovengenoemde uitdagingen te overwinnen worden in dit proefschrift doelstellingen opgezet als:

1. onderzoek naar kleurverschuivingsmechanismen voor diffusiematerialen: BPA-PC en PMMA
2. onderzoek naar kleurverschuivingsmechanismen voor belangrijke reflecterende materialen: MCPET en MCPOLYCA
3. kleurverschuivingsonderzoek voor mid-power LEDs, onderzoeksmethode voor de kwantificering van de kleurverschuiving, veroorzaakt door elk afzonderlijk onderdeel in de LED-armatuur

4. versnellingsmethode of aanpak voor de kleurverschuiving op armatuurniveau en voorspelling voor de kleurverschuiving hierin.

Kleurverschuiving voor optische materialen is onderzocht en een vergelijking tussen twee veelgebruikte diffusoren, BPA-PC en PMMA is gemaakt. In dit hoofdstuk worden veelgebruikte commerciële diffusiematerialen (BPA-PC en PMMA) experimenteel onderzocht naar de kleurverschuivingseffecten tijdens veroudering. Daarnaast worden ook kleurveranderingsmechanismen van degradatie van transmittantie bestudeerd. Resultaten zijn:

- Inconsistente afbraak van golflengte-afhankelijke transmittantie induceert de afname van de blauwe / gele lichtintensiteitsverhouding en leidt dus tot de kleurverschuiving naar het gele veld, dat is het kleurverschuivingsmechanisme van BPA-PC;
- Ook voor de niet-verouderde BPA-PC varieert de transmissie met golflengte in het zichtbare lichtveld door de chemie van het materiaal, wat de verandering van intensiveringsverhouding van blauw licht tot geel licht in de SPD veroorzaakt, waardoor kleuren veranderen in perceptie;
- Oxidatie speelt een sleutelrol bij de afbraak van de transmissie bij de piekgolflengte van het blauwe lichtveld, dat in verband staat met de verkleuring van thermische BPA-PC materialen. Echter voor het PMMA-monster tot 3000 uur, was er geen oxidatie bij 85deg.C, noch bij extra blootstelling aan blauw licht, noch zelfs bij extra vochtigheid van 85% RH.

Kleurverandering is verder onderzocht op de PMMA diffuser, conclusies zijn:

- Verkleuring werd niet waargenomen voor een monster dat is onderworpen aan veroudering op van 85 ℃ gedurende 5000 uren, of met extra blauwe lichtbestraling gedurende 5000 uren of met extra vochtigheid van 85% RH gedurende 5000 uren of zelfs bij veroudering van 100 ℃ voor 3000 uren.
- Materialen onderworpen aan veroudering op 150 ℃ gedurende 360 uren heeft een merkbare verkleuring aan de oppervlakte en heeft een significante golflengte afhankelijke afbraak in het transmissiespectrum veroorzaakt door oxidatie. Het monster met veroudering op 100 ℃ gedurende 3000 uren heeft minder oxidatie, hoewel er geen significante transmissiespectrumreductie werd waargenomen.
- Door gebruik te maken van een dergelijk model als een diffuser die op een LED-armatuur is gemonteerd, heeft de piekintensiteit van de stralingsflux in het blauwe lichtgebied een ernstiger reductie dan die in het gele lichtgebied, wat resulteert in een vermindering van de stralingsfluxintensiteit.

Verhouding van blauw licht tot geel licht derhalve zorgt ervoor dat de kleurverschuiving naar het gele spectrum gebeurt.

Degradatie van microcellulaire PET reflecterende materialen gebruikt is eveneens experimenteel bepaald. Zowel de lichtverschuiving als de kleurverschuiving zijn onderzocht. Failure modes en degradatie mechanismen werden ook onder verschillende omstandigheden onderzocht. Resultaten tonen aan dat:

- Een vochtigheidstest bij 85 ºC en 85% RH gedurende 4000 uur (of zelfs korter) kan leiden tot hydrolytische afbraak, waardoor zowel de afname van reflectie als gevolg van golflengte en een ernstige verbrossing optreedt;
- Oxidatieve afbraak na 4000 uur op 85 ºC kan leiden tot een lichte reflectiespectrumsverandering, terwijl het extra blauwe licht weinig impact heeft;
- Kleurverschuiving veroorzaakt door thermische veroudering bij 85 ºC gedurende 4000 uur is 0.0001 en de lumen efficiëntie daalde met 1,57%. Wanneer microcellulaire PET verkrummelt door de embrittlement tijdens de vochtigheidstest, stijgt de kleurverschuiving naar 0.0004 en wordt de efficiëntie van de lumen verminderd met 4.47%.

Kleurverschuivingen van veel gebruikte mid-power witte LEDs onder verschillende verouderingsomstandigheden zijn onderzocht. Het type mid-power LEDs dat is onderzocht, wordt ook veel gebruikt in downlight armaturen voor binnenverlichting toepassingen. Kleurverschuivingsmodi en mechanismen veroorzaakt door verschillende belastinging is onderzocht en een acceleratie- en voorspellingsmethode is voorgesteld. Temperatuur, vocht en stroom is experimenteel onderworpen aan de mid-power LEDs om kleurveranderingsmechanismen te onderzoeken. Conclusies kunnen worden getrokken als:

- Er isneen exponentiële relatie tussen kleurverschuiving (Δu', Δv') en verouderingstijd bijna voor alle verouderingsomstandigheden. We kunnen de kleurverschuiving Δu 'en Δv' extrapoleren op basis van de gevonden regressievergelijkingen en zo een voorspelling doen voor de totale kleurverschuiving Δu'v ';
- Stroom verzoorzaakt een andere faal mechanisme. De verandering van de piektintensiteit laat zien dat verhoogde stroom de afbraak van de LED chip versnelt;
- Vochtigheidstest veroorzaakte een aanzienlijke kleurverschuiving zowel in u' als v'. Analyse van de peakintensiteitsvergelijking laat zien dat het niet alleen om de afbraak van de LEDs gaat, maar ook de afbraak van de fosforlaag draagt bij tot een significante kleurverschuiving in de vochtigheidstest.

Om de kleurverschuiving die door elke afzonderlijke component in de LED-armatuur werd veroorzaakt, te kwantificeren, werd een nieuwe aanpak voor kleurverschuivingsonderzoek op LED-gebaseerde armaturen voorgesteld. Met behulp van deze aanpak kan de kleurverschuivingsbijdrage van elk afzonderlijk onderdeel (diffuser, reflector, behuizing en LEDs) worden berekend naar de kleurverschuiving op armatuurniveau. Deze aanpak is gebaseerd op de zogenaamde view factor methode en is toegepast op een specifiek LED-armatuur. De aanpak die in dit proefschift wordt voorgesteld, kan gemakkelijk worden gebruikt om de kleurverschuivingsbijdrage van elk afzonderlijk onderdeel voor LED-armaturen te extraheren. Volgens de resultaten van de simulatie- en berekeningsmethode, na veroudering voor 4000h, heeft de downlight een totale kleurverschuiving van 0.002, zelfs minder dan de kleurverschuiving van de afzonderlijk LEDs, die ongeveer 0.0025 bedraagt. Dit komt doordat de twee belangrijke contributors LEDs en diffuser een tegenovergestelde bijdrage hebben in de kleurverschuivingskomponent van $\Delta v'$. De component $\Delta v'$ van de LEDs heeft een afname van 0.0026, vergeleken met de toename van ongeveer 0.0017 veroorzaakt door diffuse transmissieverandering tijdens veroudering. Kleurverschuiving veroorzaakt door veroudering van LEDs is heel anders dan die van diffuser. De relatieve fluxafbraak van de LEDs in de SPD kwam duidelijk voor in het gele lichtgebied, waardoor de kleurverschuiving naar blauw werd veroorzaakt. Daarentegen verschuift de kleur tot geel voor veroudering van de diffuser, aangezien er veel meer afbraak van flux in het blauwe lichtgebied is. De totale kleurverschuiving van de armatuur is hierdoor naar blauw, en de LEDs domineren de kleurverschuivingsrichting van de LED-armatuur in dit onderzoek. De kleurverschuiving door LEDs en door diffusers (en reflectoren) worden door elkander gedeeltelijk teniet gedaan. De totale kleurverschuiving van de armatuur is daardoor zelfs minder dan de kleurverschuiving van de LEDs.

Richtlijnen en ontwerpregels zijn in dit proefschrift voorgesteld op basis van de resultaten en bevindingen in het volledig onderzoek, en zijn nuttig bij het ontwerpen of vervaardigen van LED-armaturen.

List of Publications （Selected）

Journals：

1. **Guangjun Lu**, M. Yazdan Mehr, W.D. van Driel, Xuejun Fan, Jiajie Fan, K.M.B. Jansen, G.Q. Zhang， Color shift investigations for LED secondary optical designs: Comparison between BPA-PC and PMMA, Opt. Mater. 45(2015) 37-41

2. **Guangjun Lu**, W.D. van Driel, Xuejun Fan, M. Yazdan Mehr, Jiajie Fan, K.M.B. Jansen, G.Q. Zhang Degradation of Microcellular PET Reflective Materials Used in LED-based Products, Opt. Mater. 49 (2015) 79–84

3. **Guangjun Lu**, W.D. van Driel, Xuejun Fan, M. Yazdan Mehr, Jiajie Fan, Cheng Qian, K.M.B. Jansen, G.Q. Zhang. Color shift and mechanism investigation on the PMMA diffuser used in LED based luminaires. Opt. Mater. 54 (2016) 282–287

4. **Guangjun Lu,** W.D. van Driel, Xuejun Fan, Jiajie Fan, Cheng Qian, G.Q. Zhang, Color Shift Acceleration on Mid-Power LED Packages, *Microelectronics Reliability, Accepted*

5. **Guangjun Lu,** W.D. van Driel, Xuejun Fan, Jiajie Fan, Cheng Qian, Huaiyu Ye, G.Q. Zhang, A Novel Approach of Color Shift Investigation on LED-based Luminaires, *under Journal review*

6. **Guangjun Lu,** W.D. van Driel, Xuejun Fan, Jiajie Fan, Cheng Qian, G.Q. Zhang, LED-based luminaire color shift acceleration and prediction, under *Journal review*

7. Xunjun He, Xingyu Yang, **Guangjun Lu,** Wenlong Yang, Fengmin Wu, Zhigang Yu, Jiuxing Jiang, Implementation of selective controlling electromagnetically induced transparency in terahertz graphene metamaterial, Carbon 123(2017)668-675

White Paper Contribution (for Department of Energy, USA):

Next Generation Lighting Industry Alliance, LED Systems Reliability Consortium, LED luminaire reliability: impact of color shift, April 2017

Available at: https://energy.gov/eere/ssl/downloads/led-luminaire-reliability-impact-color-shift

Book Chapter:

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1. **Guangjun Lu**, W.D. van Driel, Xuejun Fan, M. Yazdan Mehr, Jiajie Fan, Cheng Qian, G.Q. Zhang, A POF Based Breakdown Method for LED Lighting Color Shift Reliability, ChinaSSL 2015

2. **Guangjun Lu**, W.D. van Driel, Jiajie Fan, Cheng Qian, Huaiyu Ye, Xuejun Fan, G.Q. Zhang， A Novel Approach for Color Shift Investigation on LED-based Luminaires, Eurosime 2016 (On line publication, no proceedings)

3. **Guangjun Lu**, Cadmus Yuan, Xuejun Fan, G.Q. Zhang, Correlation of Activation Energy between LEDs and Luminaires in the Lumen Depreciation Test, Proceedings of the 15th international Conference on Thermal, Mechanical and Multi-Physics Simulation and Experiments in Microelectronics and Microsystems, PP. 1-3, 2014

4. **Lu GJ**, Yuan C, Tang H, Wong C, Fan XJ, Zhang GQ, Cause analysis on highly depreciated indoor LED product in CSA020, Proceedings of China Solid State Lighting Forum, 2013

5. **Lu GJ**, Yuan C, Sun B, Li B, Fan XJ, Zhang GQ, Validation of the methodology of lumen depreciation acceleration of LED lighting, Proceedings of China Solid State Lighting Forum, 2013

6. Cell K.Y. Wong, Stanley Y.Y. Leung, **Guangjun Lu**, et.al, Impact Process Parameters on Color Consistency of Mid/Low Power LED Package, Proceedings of China Solid State Lighting Forum, 2012

Acknowledgements

First, I am grateful to the people from China Solid State Lighting Alliance (CSA) and TU Delft who jointly developed the international research cooperation program in the form of Beijing Research Center. This gave me a unique opportunity to pursue my Ph.D research activities both in China and in the Netherlands. Besides, CSA offered me not only an excellent scientific platform but also broad connections with global LED industry, which facilitated my research activities. Meanwhile, I am grateful to Changzhou base of State Key Laboratory for Solid State Lighting for their financial support, experiments set-up and opportunities to have some soft skills training. This research was partially financially supported by China's 863 Program (No. 2015AA03A101), and also accomplished within EMRP JRP ENG62 MESaIL which was carried out with funding by the European Union.

This was really a wonderful journey in my life. During this journey, I have received so much help, support and encouragement from many people, and I would like to express my gratitude to every one of them. Without them, I would not have completed my thesis.

I owe my indebtedness to my promotor Prof. Dr. Guoqi (Kouchi) Zhang. I couldn't start this great journey to work as a PhD candidate without the opportunity offered by Prof. Zhang. I have gained tremendously from Prof. Zhang. He not only nurtured me in my academic field, but also impressed me in his emphasis on communication with different kinds of people, which I believe is beneficial to my whole life. Prof. Zhang encouraged me to communicate with internal and external resources, not just stay alone, or try to communicate with people I like, but more importantly to learn how to communicate with somebody who is difficult to deal with.

I would like to express my sincere thanks to my copromotor Dr. Willem Dirk Van Driel. Dr. Van Driel is so kind and always optimistic, and he works efficiently. Although he and I did not meet many times, every time our meeting and conversation are effective and efficient. He shared with me the latest research progress in the industry and his view and thinking generously. He brought new ideas and significant breakthrough to my research every time after we met. Dr. Van Driel's review on my manuscript is also efficient, and his correction and comment are so clear and accurate that it is easy for me to make a revision every time after his review.

My special thanks go to my advisor Prof. Xuejun Fan, who helped me a lot

during the four years when I was a PhD candidate especially in the first year when I was in Changzhou. Prof. Fan shared his view, and helpful publications related to my research in time and gave direct comments to me for my manuscripts, which increased efficiency and added value to my research.

I would like to extend my sincere gratitude to Dr. Maryam Mehr. Dr. Mehr provided me with great help when I was in Netherlands even when she was busy with her research at that time. She went with me to Willem in Philips lighting for project discussion and aging materials preparation and to TNO for lighting characterization, helped me with aging experiments set-up, and introduced me to scientists who performed materials analysis. More importantly, she contributed the most valuable idea in my first publication.

My sincere thanks also go to Dr. Sau Koh who helped me not only in the registration of PhD candidate, coordination between SKL and BRC, but also cared about my research especially in the first year. Dr. Sau Koh gave me some very helpful tips about living in the Netherlands and showed me around the city Hague next to Delft when I first visited Netherlands. He introduced some other senior students to me and asked them to help me in many things.

I'd like to give my thanks to Prof. Kasper Jansen. He shared his oven to me for my samples aging, gave me very helpful tips on the experiments set-up of aging conditions. He taught me how to perform stress-strain experiments. He revised my manuscripts very carefully and efficiently. In addition, he gave me some valuable suggestions on my research plan in the go-no-go meeting. Dr. Cell Wong helped me a lot during the first year when I was in Changzhou base of State Key Lab. She, as my daily supervisor, had very strict requirements on me about my research and conference publication writing, and she also gave me very direct suggestions or comments. I also want to thank Dr. Jiajie Fan and Dr. Cheng Qian, who helped me a lot in the Journals publication when I was in the SKL of Solid State Lighting.

My special thanks go to Mr. Herman Broekhuizen from the faculty of Industrial Design Engineering of TU Delft for specimen preparation and support of aging process. Mr. Broekhuizen kept regular maintenance for the aging equipment in the humidity and temperature aging tests, which lasted around one year. He is very easy to deal with and we exchanged thinking and ideas freely, and he encouraged me on my PhD research. He became my best friend when I left Delft. I cherish the friendship between us.

I also would like to express my special thanks to Prof. Jinghua Yin, my advisor in the period of master courses. She cared me very much not only on my research but also on my career development. It is she who made a

recommendation for my entrance to the TU Delft.

I'd like to give my thanks to Prof. Kees Beenakker who cared about my research and my staying in Netherlands. He is friendly and gave me encouragement no matter when we met or communicated by email. He is also the one who agreed to offer me the opportunity to become a PhD candidate.

Prof. Lina Sarro cared about research when I was in the campus of Delft. She gave me many suggestions about my research in the go-no-go meeting. I'd like to express my thanks to her. I would like to thank our secretaries Marian Roozenburg and Xandra Tober for their wonderful support.

I am grateful to Dr. Lijing Xue in the Faculty Aerospace Engineering of TU Delft for FTIR measurement and data analysis support. She not only taught me how to do analysis on the measurement results, shared her view on my results, but also gave me encouragement on my journal publication writing. I'd like to give thanks to Mr. Freddie Furrer of TNO Eindhoven for integrating sphere measurements. Mr. Furrer responded to my test requests positively, helped set up the test equipment efficiently and carefully, and taught me how to use the equipment. He is friendly and enthusiastic to me. I'd like to treat him if I got chance. Mr. Paul Debeen in Philips Lighing gave me many samples for my research, and I would like to express my gratitude to him. Hairen Tan helped me in the optical materials parameters test, not only for the test equipment booking but also for the abnormal test result analysis. He also gave me very direct suggestions on my revision of manuscript submission. I'd like to express my thanks to him. Thanks also go to Dr. Rudi Santbergen and master student Mathew Alani from EWI of TU Delft who also helped me for the optical materials measurement.

When I was in Delft, many friends or colleagues gave me help, and brought pleasure. I'd like to thank them. They are Huaiyu Ye, Jiaqi Tang, Jing Zhang, Pan Liu, Jia Wei, Teng Ma, Lin Qi, Xueming Li, Daniel Yi, Rene Poelma, StenVolberget, Xin Guo, Manjunath R. Venkatesh, Pengfei Sun and some I even didn't remember their names. I still remember that Dr. Henk van Zeijl brought us much fun in the canoeing in Delft.

BRC colleagues and friends like Jianlin Huang, Minzhi Dong, Yang Liu, Hongyu Tang and Hao Zhang, Yuan Gao, etc, gave me much help. Also some contributed to my PhD research located in Changzhou, they are Liangliang Luo, Shuting Gao, Jie Zhuang, Gongqi Fan, Min Jia, Yongqiao Qin, Yanjian Xiong, Lei Li, Lei Zhong, Kai Lin and Zhike Zhu. I'd like to give my thanks to them. Meng Pan, Gaojin Qi, Wei Zhang and Yong Yu, we enjoyed much good time in Changzhou. It's interesting and memorable that I obtained the drive license

during the period. Mingxing Zhu and Haining Wu are good partners in driving excises or test and we had a lot of fun.

I got much support and encouragement from my previous colleagues and friends during the Ph.D study. Mr. Jim Qi is my previous colleague in GE Healthcare and become one of my best friends. We have very similar thinking and interests in many things. He has superb talent in English and helped me a lot in my English writing. He is also a poet and has a wonderful sense of humor, which added much fun to my academic life. He once arranged a special transit in Amsterdam to see me when he went back to Canada from China. Lei Wang, another previous colleague and good friend in GE Healthcare, cared about my research and career development, and even introduced me to the hiring manager for new job opportunities in GE Healthcare. Srini, my previous boss and mentor in GE Healthcare, cared about me and sent emails to me for my birthday celebration in 4 consecutive years. Zhiping Hu, William Wang, Edward Zhou in Fairchild Semiconductor now On Semiconductor, also gave me much encouragement. Bruce Huang, my previous colleague and friend in Freescale (Motorola), now in Apple, cared about my research progress and career. I'd like to give my sincere thanks to them.

Finally and most importantly, I would like to express my uttermost thankfulness to my dear family, my father and my mother, my two older brothers and my younger sister, my sisters-in-law, my nephews and my niece, my uncles and my aunts. Your endless love is the most powerful source not only in this thesis but also in my life.

About the Author

Guangjun Lu was born in 1980, in Hubei Province, China. He commenced in 2002 from Harbin University of Science and Technology with a Bachelor of Science (BS) in Electronics Materials and Devices. He then continued to read for his Master of Science (MSc) in Materials Physics and Chemistry from Harbin University of Science and Technology after his commencement. In 2005, he received his MSc. and continued his research and teaching career in Harbin University of Science and Technology. After one year of successful teaching and research in the same university, he obtained an opportunity to serve Freescale Semiconductor (originated from Motorola) (Tianjin) as a process and package development engineer in the department of Global Package Engineering (GPE) for automotive devices, where he played a role in solving a well-known solder void issue. Then, in 2008 he broadened his career development opportunity in Fairchid Semiconductor (Suzhou) as a senior product engineer covering smart power module (spm) products in the department of Asia Pacific Product Engieering (APPE), where he was leading many projects and activities including new product introduction, yield improvement, cost reduction and customer complaint handling. It was during this period he was successfully certified as Six Sigma Black Belt in recognition of his accomplishment of two important yield improvement projects. Then, in 2011, he furthered his role of full-time six sigma black belt (leadership professional band) in the department of Advanced Manufacturing Engineering (AME) GE Healthcare, based in Wuxi, where he contributed a leading role in several DMAIC projects. Although working in GE Healthcare not very long, Mr. Guangjun Lu won the GE Healthcare Global best practice and the yearly quality award of 2012 thanks to the substantial yield improvement of one key Ultrasound product in GE Healthcare.

In November 2012, Mr Guangjun Lu decided to conduct his Ph.D research in the area of the LED based luminaire color shift and aimed to understand the color shift mechanisms, quantify the color shift contribution by each individual component in a luminaire and propose an acceleration and prediction method/approach for color shift of luminaire level products at Beijing Research Center, TU Delft. Guangjun Lu is now working as associate director for lean sigma operations in Wuxi Apptec, a leading global pharmaceutical,

biopharmaceutical, and medical device open-access capability and technology platform with global operations designed to help worldwide customers shorten the discovery and development time and lower the cost of drug and medical device R&D through cost-effective and efficient solutions.