

Light-diffusing materials for LED illumination applications: Comparing the effectiveness of two typical light-diffusing agents

Xing Ouyang, Peibin Li, Dazhu Chen, Jiaoning Tang

Shenzhen Key Laboratory of Special Functional Materials, College of Materials Science and Engineering, Shenzhen University, Shenzhen, 518060, People's Republic of China

Correspondence to: X. Ouyang (E-mail: oyx@szu.edu.cn) or Dazhu Chen (E-mail: dzchen@szu.edu.cn)

ABSTRACT: Light-diffusing materials (LDMs) are the primary lampshade materials used for indoor light-emitting diode (LED) illumination. In this work, two varieties of typical LDMs filled with acrylic or silicone diffusing agents were fabricated via a twin-screw extruder, and the influences of the particle size, refractive index and light-diffusing surface area of the light-diffusing agents on the properties of the LDMs, including their light-diffusing properties, thermal stability and mechanical properties, were explored in depth. The results indicated that both acrylic and silicone light-diffusing agents can be dispersed evenly in a polycarbonate (PC) matrix. The light-diffusing surface area and refractive index are the main factors that influence the diffusing properties of LDMs. Compared with the silicone LDM, the acrylic LDM exhibited higher transmittance but poorer thermal stability and impact strength, making this material suitable for a rapid injection molding process to form half-sphere covers that combine high haze and transmittance for LED light bulbs. Silicone LDMs exhibit good stability and impact strength, favoring a slow single-screw extrusion molding process to prepare tubular or panel covers for tubular LED lamps and panel lamps. In this study, a method for producing LDMs with both high transmittance and high haze is reported. © 2015 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2016**, *133*, 42923.

KEYWORDS: composites; morphology; optical properties; polycarbonates

Received 2 April 2015; accepted 7 September 2015

DOI: 10.1002/app.42923

INTRODUCTION

Light-diffusing materials (LDMs), which can diffuse light via Rayleigh scattering, were first used in liquid crystal displays^{1–3} because they can spread light from a point source and produce uniform luminance. Recently, LDMs were observed to be able to transform the intense light from an LED source into illumination that is both uniform and soft (Figure 1); as a result, LDMs are becoming key lampshade materials for indoor LED illumination.

Transmittance and haze are important light-diffusing properties of LDMs. These materials are expected to exhibit good light-diffusing properties, i.e., high transmittance combined with high haze, because high haze can make luminance more uniform and high transmittance can improve brightness. However, transmittance and haze are necessarily conflicting properties because high haze, which results from light scattering, often decreases transmittance. Obtaining high transmittance combined with high haze is a significant challenge related to LDM technology.⁴ Decreased transmittance is attributed to strong backward scattering. High haze combined with high transmittance can be attained by reducing backward scattering and

strengthening forward scattering. To achieve this objective, researchers have used many approaches,^{5–7} including roller embossing,⁶ the deposition of micrometer dots onto a polymer film surface,⁸ and a hybrid extrusion roller embossing process,⁵ to prepare LDMs. A diffusing film with excellent diffusing properties can be prepared using these methods; however, the technical conditions are difficult and the methods are only appropriate for the fabrication of flat plates. Therefore, the widely used alternative method is to disperse microspheres, i.e., so-called light-diffusing agents, into a transparent polymer resin via a mixing process.^{1–4,9,10} In the composites prepared by this method, the diameter and relative refractive index of the diffusing agent are critical for the light-diffusing properties.^{1,2,11–13} If a light-diffusing agent with suitable diameter and relative refractive index is selected, the forward scattering of the LDM will be increased, resulting in both high haze and high transmittance.

Many substances, such as silica, barium sulfate, polystyrene, polyacrylic, and silicone, can be used as light-diffusing agents when their shapes are microspherical.^{1–4,9,10} Among these substances, acrylic and silicone microspheres remain the preferred materials for use in LDMs for indoor LED illumination^{1,2} because of their desirable light-diffusing properties. To our

knowledge, although these two types of light-diffusing agents are extensively used in LDMs for LED illumination, a systematic study of their characteristics has not yet been reported in the literature, and these agents have not yet been subjected to a detailed comparison regarding the influence of their refractive index, particle size, and surface area on their light-diffusing properties and thermal stabilities.

Generally, polycarbonate (PC) is preferentially selected as the polymer matrix in which the light-diffusing agents are dispersed to prepare LDMs because PC exhibits good mechanical properties and can be molded into a tubular shape by extrusion or into a semispherical shape by injection. In this work, LDMs were prepared by dispersing acrylic or silicone light-diffusing agents into a PC matrix through a melt extrusion mixing process. The influences of the particle diameter and refractive index on the light-diffusing properties and thermal stabilities of the corresponding LDMs were studied in depth. A method for producing LDMs with both high transmittance and high haze is reported.

EXPERIMENTAL

Materials

Two types of light-diffusing agents—acrylic and silicone—were used in this work. The acrylic light-diffusing agent, BMSA, has a specific gravity of 1.1 g/cm³ and was purchased from Sekisui Plastics Corp., Ltd. The silicone light-diffusing agent X-52-7056A has a specific gravity of 1.3 g/cm³ and was purchased from Shin-Etsu Chemical Corp., Ltd. These two types of commercial light diffusing agents are widely used in LDM production. The main components of BMSA and X-52-7056A are crosslinked polymethyl methacrylate and polymethyl silsesquioxane, respectively, and the corresponding refractive index values determined using an ATAGO NAR-1T Abbe refractometer via the powder method¹⁴ are 1.484 and 1.372, respectively. The polycarbonate (PC) resin, CALIBRE 201-10, exhibited a melt flow index of 10 and was purchased from DOW Chemical Corp., Ltd.; this resin was selected as the base material because of its excellent mechanical properties. The refractive index of the resin was measured to be 1.581. The pentaerythritol stearate used as a lubricating agent and the tris(2,4-di-*tert*-butyl) phosphite and *n*-octadecyl- β -(4-hydroxy-3,5-di-*tert*-butylphenyl) propionate used as antioxidants in this study are common additives for PC modification. All materials were used as received, without further treatment.

Fabrication of LDMs

The PC pellets and light-diffusing agents were compounded using a Coperion Corp. extruder (model ZSK 27) with a 27-mm corotating intermeshing twin screw containing 10 mixing zones. The length/diameter ratio of the extruder was 40. The aforementioned screw design was chosen to obtain a low shear force and to guarantee the minimum degree of PC degradation and good dispersion of the light-diffusing agent in the polymer. The temperature sequence from the hopper to the die was 200, 240, 240, 245, 245, 245, 245, 245, 245, and 245°C. The screw speed was 300 rpm. Polymer pellets, the light-diffusing agent and other additives were first mixed and then introduced into Zone 1. After passing through the extruder, the polymer strands

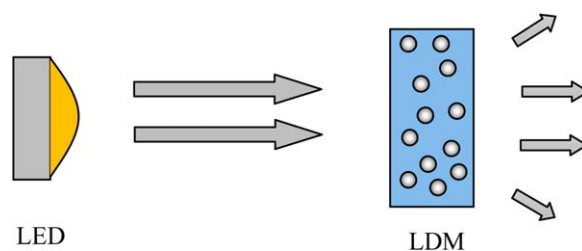


Figure 1. Schematic of LDM function. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

(3 mm in diameter) first entered a water bath and were then cut by a pelletizer to produce ~3-mm-long pellets. After being compounded, the pelletized composite resin was dried and then stored in a moisture-barrier bag prior to injection molding.

ASTM Type I tensile bars and notched impact test specimens with a thickness of 3.2 mm and 30-cm-diameter disks with a thickness of 1 or 2 mm were fabricated using an LK injection-molding machine (model PT80) with a 40-mm-diameter single screw and a length/diameter ratio of 18. The lengths of the feed, compression, and metering sections of the single screw were 396, 180, and 144 mm, respectively.

Characterization

The morphologies of the light-diffusing agents and LDM fracture surface were observed using a scanning electron microscope (Hitachi SU-70). A drop of dilute microsphere dispersion in EtOH was spread onto a glass surface and dried in a dust-free environment at room temperature. Prior to observation, both the dried microsphere and LDM fracture surface were coated with a thin layer of gold under vacuum.

The particle size and size distribution were determined by measuring at least 200 microspheres from the SEM photographs using an Mshot image analysis system. The number-averaged diameter (D_n), weight-averaged diameter (D_w), and polydispersity index (PDI) were calculated according to eqs. (1–3), respectively. The specific surface areas of microspheres (S) were calculated using eq. (4).

$$D_n = \frac{\sum i n_i D_i}{\sum i n_i} \quad (1)$$

$$D_w = \frac{\sum i n_i D_i^4}{\sum i n_i D_i^3} \quad (2)$$

$$PDI = \frac{D_w}{D_n} \quad (3)$$

$$S = \frac{6 \sum i n_i D_i^2}{\sum i n_i \rho D_i^3} \quad (4)$$

where D_i and n_i are the diameter and number of particle i , respectively, and ρ is the specific gravity of the light-diffusing agent.

Light-diffusing properties, such as transmittance (T) and haze (H), were tested using a light-diffusion system (EEL 57D) according to ASTM D1003. The light source used in this apparatus was a halogen tungsten lamp. The values of T and H were calculated from eqs. (5) and (6), respectively:

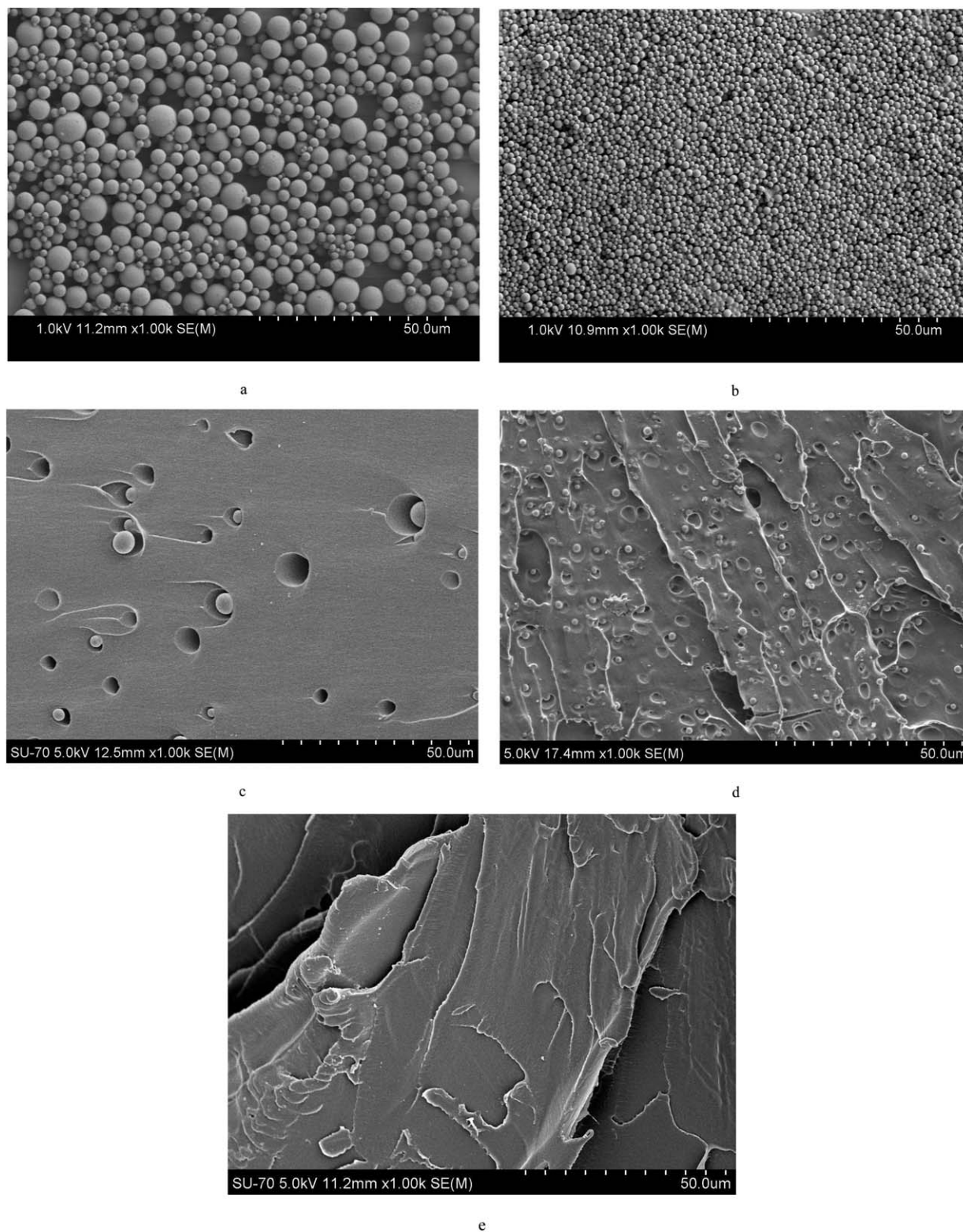


Figure 2. SEM images of light-diffusing agents and LDM fracture surfaces. (a) acrylic particles, (b) silicone particles, (c) acrylic LDM containing 1 phr of light-diffusing agent, (d) silicone LDM containing 1 phr of light-diffusing agent, (e) neat PC.

$$T = \frac{I_D + I_R}{I_0} \quad (5)$$

$$H = \frac{I_D}{I_D + I_R} \quad (6)$$

Table I. Particle Size and Specific Surface Area of Two Types of Light-Diffusing Agents

Samples	D_n (μm)	D_w (μm)	PDI	S (cm^2/g)
Acrylic type	4.22	4.85	1.15	11735
Silicone type	2.09	2.19	1.05	22465

where I_0 , I_D , and I_R are the intensities of the incident light, diffuse transmission and regular transmission, respectively.

The thermal stability was tested using a TA Instruments thermogravimetric analyzer (TGA 50) under nitrogen protection at a heating rate of $10^\circ\text{C}/\text{min}$ over a temperature range of 30 to 700°C . The thermal decomposition activation energy was calculated using the Freeman-Carroll method.¹⁵

The tensile properties of the LDMs were measured using a universal testing machine (CMT6104, MTS Systems Corp.) according to ASTM D638. The tensile speed and gauge length were 2 mm/min and 20 mm, respectively. The Izod impact strength was measured on notched specimens using an impact tester (ZBC7000, MTS Systems Corp.) according to ASTM D256. The impact speed was 3.46 mm/s. All mechanical measurements were performed at room temperature. For each composite, five specimens were tested, and the average value was recorded.

RESULTS AND DISCUSSION

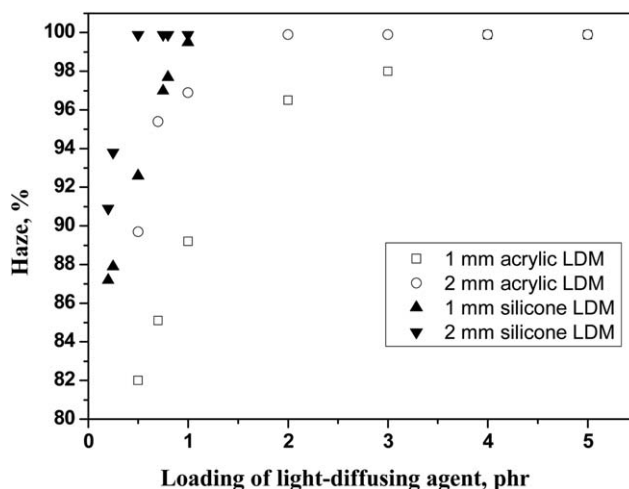
Dispersion of Microspheres in the Matrix

Figure 2 shows the morphologies of acrylic-type and silicone-type light-diffusing agents and the fracture surfaces of the LDMs. These two types of light-diffusing agents are both regularly spherical, and their mean particle size, particle size distribution, and specific surface area were analyzed from Figure 2(a,b) using the Mshot image analysis system. The results calculated according to eqs. (1–4) are listed in Table I. As shown in Table I, the silicone particles have smaller particle diameters and a narrower size distribution than the acrylic particles.

Compared with the flat fracture surface of neat PC, the LDM fracture surfaces contain many microbeads and microholes. The sizes of the micro-holes on the acrylic LDM fracture surface are larger than those on the silicone LDM fracture surface, which is consistent with the diameters of the light-diffusing agents. In addition, as evident in the graphs, both types of light-diffusing agents were uniformly dispersed in the PC matrix.

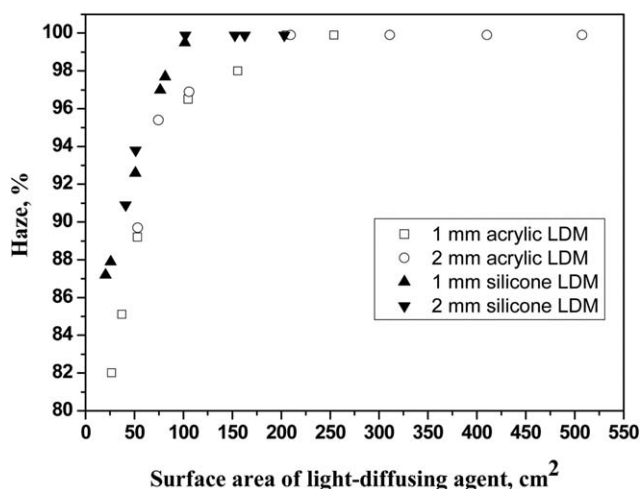
Light-Diffusing Properties of LDMs

Figure 3 shows that the haze produced by LDMs increases to a maximum of 99.9% as the loading of the light-diffusing agent increases. For a 1-mm-thick silicone LDM plate, the haze reaches a maximum when the loading of the diffusing agent is 1 part per hundred parts of resin (phr), whereas the haze is maximized with a loading of only 0.5 phr for a 2-mm-thick silicone LDM plate. For a 1-mm-thick acrylic LDM plate, the haze reaches a maximum of 99.9% at an agent loading of 4 phr, and for a 2-mm-thick acrylic LDM plate, the maximum haze is achieved at a loading of 2 phr. As shown in Figure 4, plates with different thicknesses reach maximum haze at a surface area of 100 cm^2 for the silicone microsphere-filled LDM plate and at

**Figure 3.** Haze vs. weight percentage of two types of diffusing agents in light-diffusing plates of different thicknesses.

200 cm^2 for the acrylic microsphere-filled LDM plate. The phenomenon of LDMs achieving maximal haze at a specific threshold of total light-diffusing particle surface area has also been reported.^{11–13} According to the literature,^{12,13} the diffusing properties are determined by the surface area of the light-diffusing agents and are less related to the particle size or size distribution, although the particle size and size distribution of light-diffusing particles are important factors that influence the diffusing properties of LDMs. Additionally, when a specific type of light-diffusing agent is used, LDMs with the same light-diffusing particle surface area will exhibit the same haze and transmittance, although the particles may have different particle sizes or particle size distributions.^{11–13}

The difference in the refractive indices of the light-diffusing agent and matrix in LDMs is another important factor that determines the diffusing properties of these materials. As the refractive index difference increases, the amount of scattered light increases; as a result, a smaller diffusing surface area is required to achieve maximal haze. The refractive indices of the

**Figure 4.** Haze vs. total surface area of various diffusing agents in light-diffusing plates of different thicknesses.

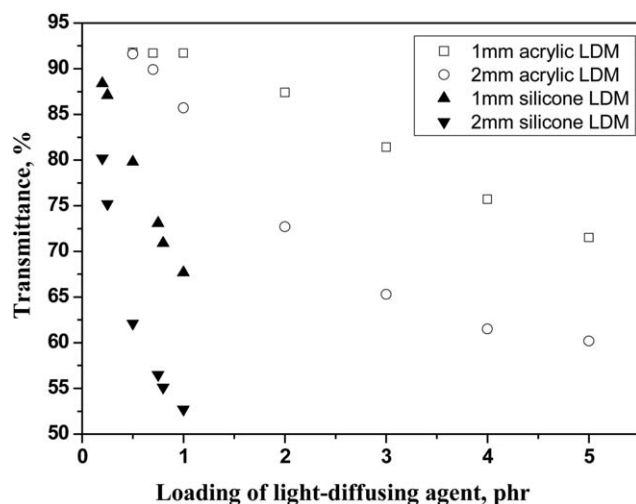


Figure 5. Transmittance vs. weight percentage of two types of diffusing agents in light-diffusing plates of different thicknesses.

neat PC and the silicone and acrylic light-diffusing agents used in this work are 1.581, 1.372, and 1.484, respectively. The difference in the refractive indices of the silicone light-diffusing agent and PC is larger than that between the PC and acrylic agent. Therefore, the loading or diffusing surface area of the silicone light-diffusing agent required to achieve maximal haze is considerably smaller than that required for the acrylic agent.

As shown in Figure 5, the transmittance of an LDM decreases as the loading of the light-diffusing agent increases; various profiles for different thickness LDM plates were examined with the same type of light-diffusing agent. Figure 6 shows that the transmittance also decreases with increasing light-diffusing surface area. The transmittance-surface area profiles for LDMs with different thicknesses also exhibit the same haze-surface area evolution profile for both types of light-diffusing agents; the influence of the light-diffusing agent loading is excluded because the diffusing properties are determined by the surface area and refractive index.

To further compare the diffusing properties of the two types of LDMs, we plotted the haze and transmittance against the surface area, as shown in Figure 7. Although both types of LDMs reached a maximum haze of 99.9% when the diffusing surface area of the light-diffusing agent achieved a critical value (100 cm² for the silicone agent and 200 cm² for the acrylic agent), the transmittances of the two types of LDMs differed: 75.7% for the acrylic LDM and 62.1% for the silicone LDM. As shown in Figure 7(a,b), for both 1-mm- and 2-mm-thick LDM plates, the acrylic LDM has higher transmittance than the silicone LDM for a given level of haze. This result is explained by the difference in the refractive indices of the two types of light-diffusing agents. Transmittance is the ratio of the intensities of the transmitted and incident light, whereas haze is the intensity ratio of the diffuse transmitted light and transmitted light. According to Fresnel's law, for a beam of vertically incident light, reflectance R , which is the ratio of reflected light to incident light, and transmittance T can be expressed using eqs. (7) and (8), respectively:

$$R = \left(\frac{n_1 - n_2}{n_1 + n_2} \right)^2 \quad (7)$$

$$T = 1 - R = \frac{4n_1 n_2}{(n_1 + n_2)^2} \quad (8)$$

Equations (7) and (8) indicate that haze has no direct relationship with R and that R increases and T decreases with increases in the refractive index difference between the light-diffusing agent and matrix. Hence, even when the haze is identical, the transmittance can be different; this phenomenon is the basis of the design of LDMs with both high haze and high transmittance. Because the difference in the refractive indices between the silicone agent and matrix is greater than that between the acrylic agent and matrix and because a greater refractive index difference increases the amount of backward scattered light and decreases the transmittance, silicone LDMs exhibit reduced transmittance compared with acrylic LDMs. Therefore, the key approach to obtaining LDMs with both high transmittance and high haze is to select a light-diffusing agent with a refractive index approximately equal to the refractive index of the matrix. This approach has practical significance for LED illumination in the context of energy conservation.

Thermal Stability of the LDMs

For high-performance PC-based LDMs for LED illumination applications, the light-diffusing agents should exhibit good thermal stability and have a suitable refractive index. Figure 8 presents the thermogravimetric curves of the light-diffusing agents and LDMs, and Table II presents the relevant parameters. Figure 8 and Table II illustrate that the silicone light-diffusing agent exhibits good thermal stability. However, for the acrylic light-diffusing agent, the decomposition onset temperature is only 150°C, and, at a temperature of 430°C, the diffusing agent is considered to have undergone complete decomposition. Compared with neat PC, whose decomposition onset temperature is 447°C, the acrylic light-diffusing agent in the acrylic-filled LDMs is decomposed completely before the PC starts to decompose, indicating that the acrylic light-diffusing agent lowers the thermal stability of the LDMs. As a result, as shown in Figure 8,

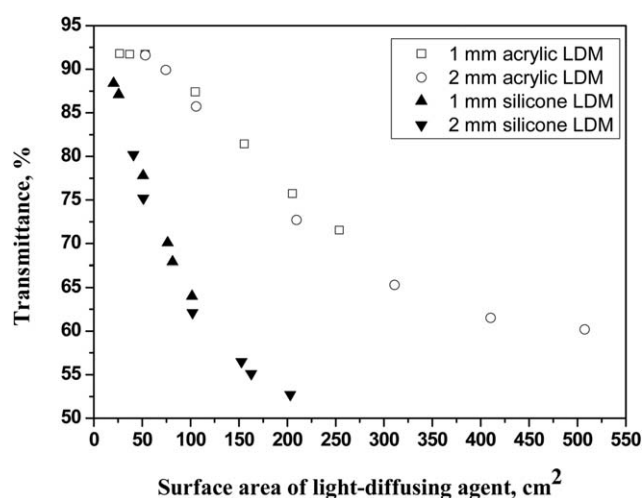


Figure 6. Transmittance vs. total surface area of various diffusing agents in light-diffusing plates of different thicknesses.

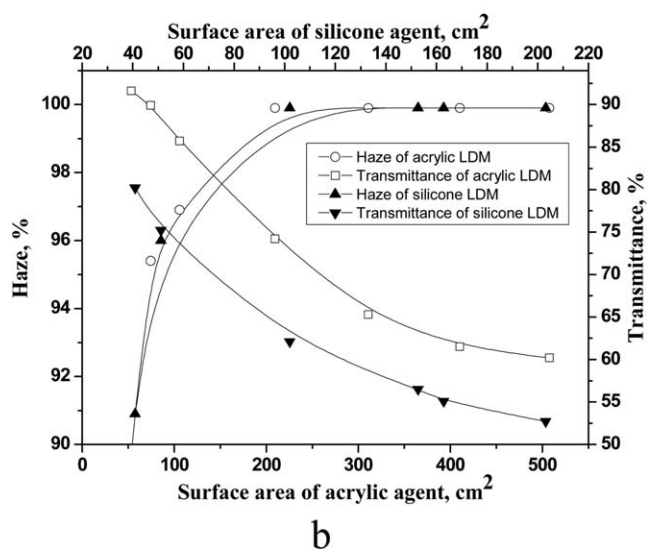
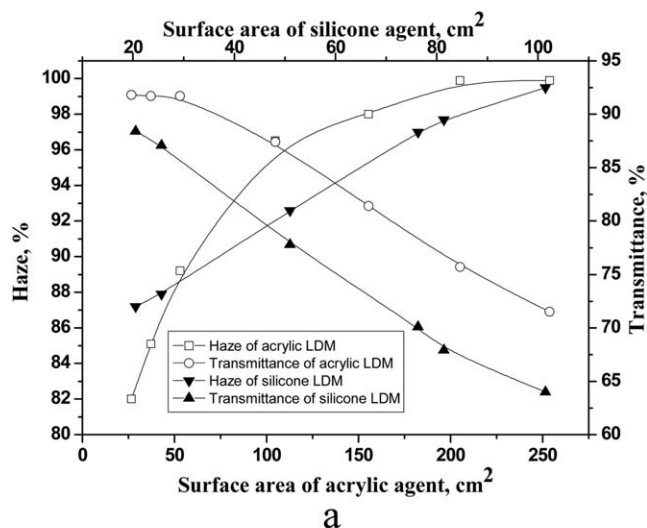


Figure 7. Comparison of the diffusion performance of the two diffusing agents. (a) 1-mm-thick light-diffusing plate, (b) 2-mm-thick light-diffusing plate.

the decomposition onset temperature of the LDM containing the acrylic light-diffusing agent at a loading of 4 phr is 408°C (remarkably lower than that of neat PC), and the decomposition termination temperature is 513°C. For the LDM containing 1 phr of silicone light-diffusing agent, the temperatures at which decomposition begins and terminates are 445°C and 517°C, respectively, which are similar to those of neat PC. Thus, the silicone light-diffusing agent adversely affects the decomposition temperature of LDMs only slightly.

The thermal decomposition activation energy is another parameter that can be used to evaluate a material's thermal stability. The Freeman-Carroll method¹⁵ was used to calculate the thermal decomposition activation energy in a N₂ atmosphere. After selecting an appropriate temperature interval to produce identical values of $\Delta(1/T)$, a line is obtained by plotting $\Delta \ln(-dW/dT)$ vs. $\Delta \ln W$, where T is the decomposition temperature in Kelvin and W is the weight percent that equals the weight of residual activated substance divided by the total activated substance in the thermal

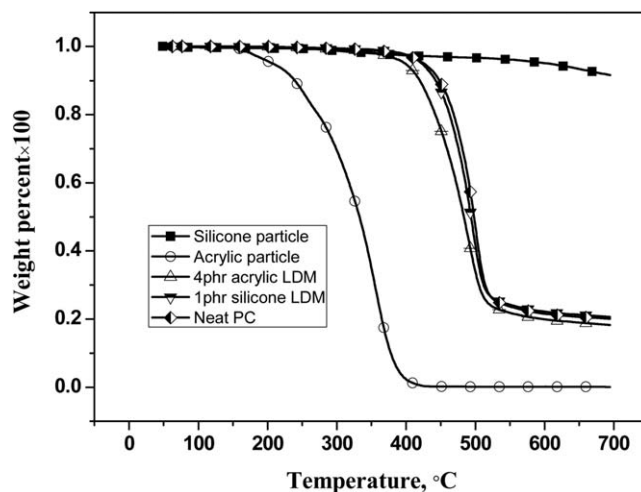


Figure 8. Thermogravimetric curves of light-diffusing agents and LDMs.

decomposition reaction. Plots of $\Delta \ln(-dW/dT)$ vs. $\Delta \ln W$ for neat PC and LDMs are presented in Figure 9. The thermal decomposition activation energy (E) is calculated via the intercept of the fitting line. The values of E are 92.45, 92.06, and 104.63 kJ/mol for neat PC, the acrylic LDM and the silicone LDM, respectively. The acrylic light-diffusing agent has a smaller influence on the thermal decomposition activation energy of the LDM, although it can lower the onset decomposition temperature substantially, as shown in the thermogravimetric curve in Figure 8. The temperature range for calculating the thermal decomposition activation energy extends from the decomposition onset temperature to the decomposition termination temperature. At temperatures lower than the thermolysis range of acrylic LDM, the acrylic light-diffusing agent is nearly completely decomposed; as a result, it has only a slight influence on the thermal decomposition activation energy of the LDM. Moreover, the silicone light-diffusing agent improves the thermal decomposition activation energy of LDM compared to that of neat PC, although it undergoes a smaller change at the decomposition onset temperature compared to neat PC. The improvement of the silicone LDM's thermal stability is attributed to the good intrinsic thermal stability of the silicone agent, in

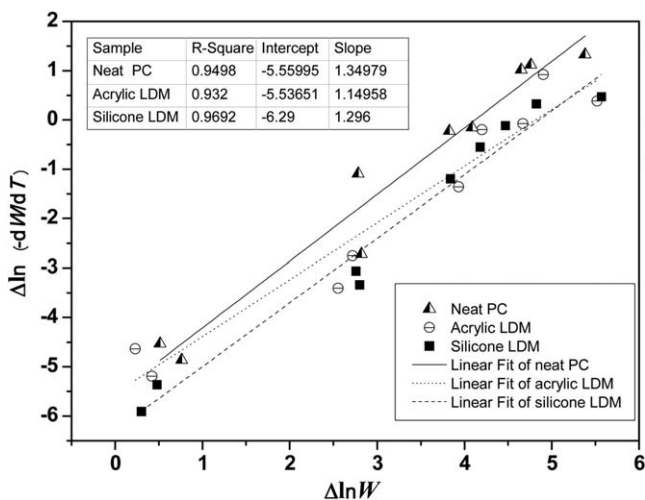


Figure 9. Thermal degradation kinetics of LDMs.

Table II. Thermogravimetric Analysis Parameters of Light-Diffusing Agents and LDMs

	Acrylic particles	Silicone particles	Neat PC	Acrylic LDM	Silicone LDM
Onset temperature (°C)	150	500	447	408	445
Termination temperature (°C)	430	690	517	513	517
Char yield at 690°C (%)	0.2 ^a	92.0	20.1	18.4	20.6

^aChar yield at 430°C.

agreement with the previously reported thermal stability results for polymer/inorganic particle composites.^{16–19}

The thermal stability of LDMs is important for their subsequent application. Because of its good thermal stability, the silicone LDM is amenable to being formed into a tubular or panel cover for a tubular LED lamp or panel lamp through a single-screw extrusion molding process at a relatively slow speed. Acrylic LDM, which is less thermally stable than silicone LDM, is not suitable for this process because the thermal decomposition of the light-diffusing agents during the long production period

may lead to a yellow stain in the final products. Although acrylic LDM lacks the good thermal stability of silicone LDM, this material combines high transmittance with high haze and can be used to produce half-sphere covers for LED light bulbs through a rapid injection molding process because the light-diffusing agent does not have the opportunity to decompose before the process is complete.

Mechanical Properties of LDMs

As shown in Figure 10(b–d), both types of LDMs are observed to maintain the good tensile strength, elongation at break and

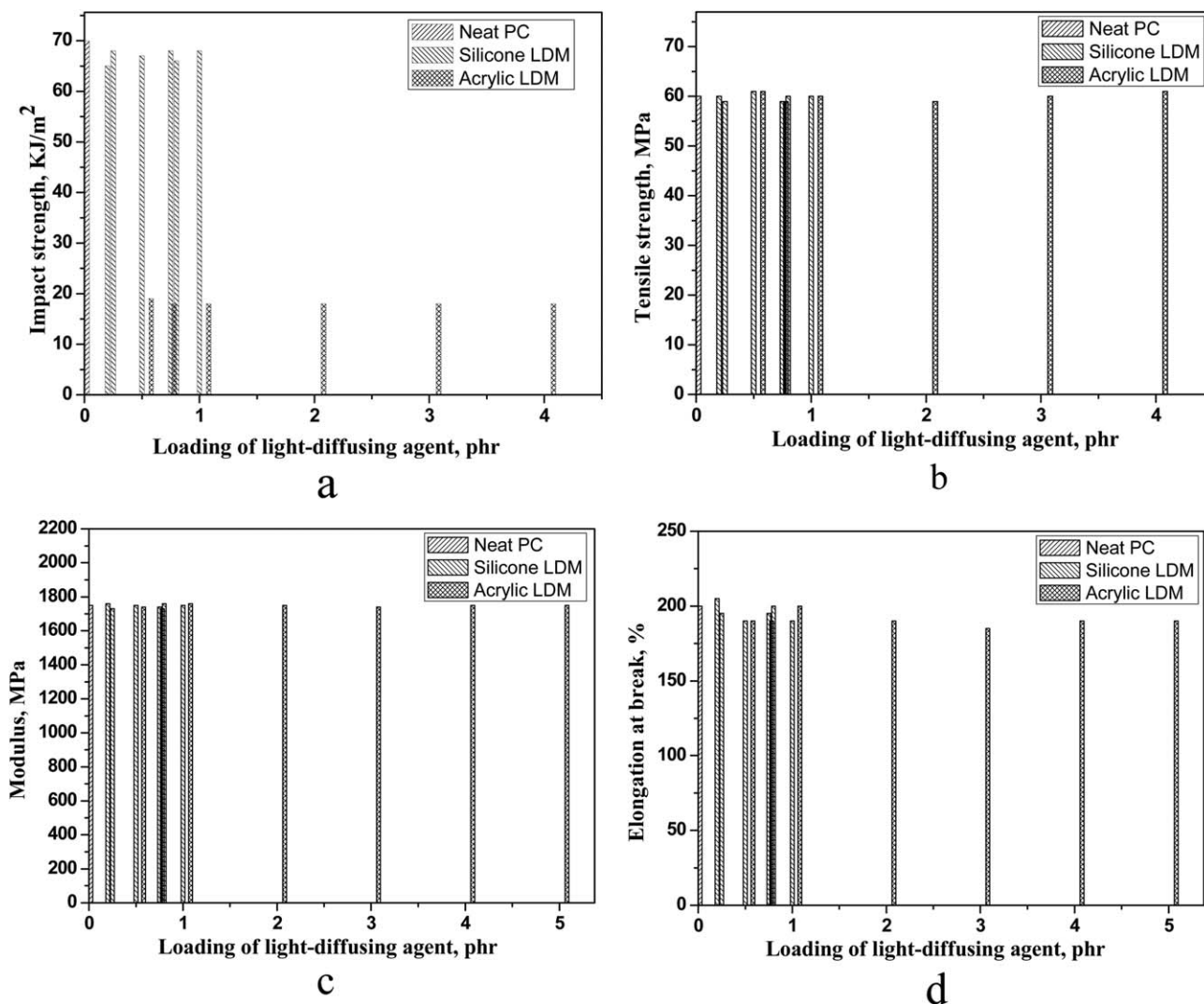


Figure 10. Mechanical properties of LDMs. (a) impact strength, (b) tensile strength, (c) modulus, (d) elongation at break.

elastic modulus of neat PC. However, compared with neat PC, the impact strength of the acrylic LDM is lower, whereas that of silicone LDM is comparable. This phenomenon is explained by considering the size difference of the light-diffusing agents. The diameter of the acrylic light-diffusing agent is 2–6 μm , whereas the diameter of the silicone light-diffusing agent is approximately 2 μm . For filled polymer composites, a smaller filler diameter corresponds to higher impact strength. Similar results were reported by Liu²⁰ for CaCO_3 -filled high-density polyethylene composites and by Kwon²¹ for spherical silica particulate-reinforced epoxy composites. The reason for this behavior might be that the defects produced by large-diameter particles decrease the impact strength. This study indicates that a silicone LDM with small-diameter particles is suitable for producing tubular and panel covers for tubular LED lamps and panel lamps due to its high strength.

CONCLUSIONS

Two types of LDMs filled with acrylic or silicone diffusing agents were fabricated via a twin-screw extruder. The acrylic and silicone light-diffusing agents were both evenly dispersed in the PC matrix. The surface area of the light-diffusing agent and the refractive index were determined to be the main factors that influence the diffusing properties of the LDMs. The maximum haze was achieved at a constant light-diffusing agent surface area for both types of LDMs. For materials that exhibit the same haze, the acrylic LDM has higher transmittance than the silicone LDM because of its lower relative refractive index compared to the PC matrix. Compared with the silicone LDM, the acrylic LDM is less thermally stable, making it suitable for a rapid injection molding process to produce a half-sphere cover for a LED light bulb that combines high haze with high transmittance. Because of the greater heat resistance and smaller particle size of the silicone light-diffusing agent, the silicone LDM exhibits good stability and impact strength, making the slow single-screw extrusion molding process to produce tubular and panel covers for tubular LED lamps and panel lamps, respectively, more appropriate. One direction for the development of new high-performance LDMs is to use a light-diffusing agent that has a refractive index similar to that of the matrix, good thermal stability, and a diameter of 1–2 μm .

ACKNOWLEDGMENTS

This work was supported by the Application Technology Innovation Project of Shenzhen University (grant number 201220) and

the National Natural Science Foundation of China (grant number 51173109).

REFERENCES

1. Ohtsuka, Y.; Fujiguchi, T.; Oishi, K. U.S. Pat. 53,52,747, October 4, 1994.
2. Ohtsuka, Y.; Fujiguchi, T.; Oishi, K. Eur. Pat. 0604130 B1; June 29, 1994.
3. Kobayashi, S.; Mikoshiba, S.; Lim, S. In LCD Backlights; Wiley: New York, 2009; Chapter 20, p 251.
4. Kuo, H. P.; Chuang, M. Y.; Lin, C. C. *Powder Technol.* 2009, 192, 116.
5. Huang, T. C.; Ciou, J. R.; Huang, P. H.; Hsieh, K. H.; Yang, S. Y. *Opt. Express* 2008, 16, 440.
6. Lu, Z. Z. J.; Lee, III, J. W.; Kim, J. M. *J. Soc. Inf. Display* 2007, 15, 565.
7. Takei, S.; Mochiduki, K.; Kubo, N.; Yokoyama, Y. *Appl. Phys. Lett.* 2012, 100, 263108.
8. Kim, G. H.; Park, J. H. *Appl. Phys. A.* 2006, 86, 347.
9. Kim, G. H. *Eur. Polym. J.* 2005, 41, 1729.
10. Kim, G. H.; Kim, W. J.; Kim, S. M.; Son, J. G. *Displays* 2005, 26, 37.
11. Chen, C. W.; Chen, C. Y. *J. Soc. Inf. Display* 2007, 15, 845.
12. Chen, C. W.; Chen, C. Y. *Colloid Polym. Sci.* 2009, 287, 1377.
13. Chen, C. W.; Chen, C. Y.; Lin, C. L. *J. Polym. Res.* 2010, 18, 587.
14. Abbe Refractometer NAR-1T Solid Instruction Manual, Cat. No.1212, ATAGO CO. LTD.
15. Freeman, E. S.; Carroll, B. *J. Phys. Chem.* 1958, 62, 394.
16. Yu, H.; Ren, W.; Zhang, Y. *J. Appl. Polym. Sci.* 2009, 113, 17.
17. Guo, B. C.; Jia, D. M.; Cai, C. G. *Eur. Polym. J.* 2004, 40, 1743.
18. Chen, G. X.; Yoon, J. S. *J. Polym. Sci. Part B Polym. Phys.* 2005, 43, 478.
19. Esthappan, S. K.; Nair, A. B.; Joseph, R. *Compos. B* 2015, 69, 145.
20. Liu, Z. H.; Kowk, K. W.; Li, R. K. Y.; Choy, C. L. *Polymer* 2002, 43, 2501.
21. Kwon, S. C.; Adachi, T.; Araki, W.; Yamaji, A. *Compos. B* 2008, 39, 740.