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Novel high performance scattering materials for use in energy saving light fittings and skylights based on polymer pigmented with polymer

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ABSTRACT

Simple quantitative performance criteria are developed for translucent materials in terms of hemispherical visible transmittance, and angular spread of transmitted luminance using a half angle. Criteria are linked to applications in luminaires and skylights with emphasis on maximising visible throughput while minimising glare. These basic criteria are also extended to angle of incidence changes which are substantial. Example data is provided showing that acrylic pigmented with spherical polymer particles can have total hemisphercial transmittance with weak thickness dependence, which is better than clear sheet, while the spread of transmitted light is quite thickness sensitive and occurs over wider angles than inorganic pigments. This combination means significantly fewer lamps can achieve specified lux levels with low glare, and smaller skylights can provide higher more uniform daylight illuminance.

1. INTRODUCTION

Pigmented polymers have been used for dispersing lamplight and daylight more or less since the widespread adoption of polymer luminaire covers and polymer skylights, some 40 to 50 years ago. What is not well appreciated is their potential for adding strongly to energy efficiency if performance is improved. Their main purpose is to disperse the transmitted radiation to produce more even illumination and to reduce the visual discomfort of direct glare from the light source itself or from the sky. Additional benefits in the case of skylights come from reducing the temporal impact of the movement of the sun across the sky on the internal distribution of illuminance. Modelling and use of these materials in design has often been approximate due to lack of suitable data on transmittance distributions. If any are available they are usually for normal incidence but output distributions change substantially with angle of incidence and also direction of incidence if there is assymetry in scattering or dispersion. It is thus in principle a demanding task to experimentally establish these distributions which are usually called the BRTF (Bi-directional transmittance and reflectance function) or BTDF with DF = " distribution function". A systematic approach to modelling and data collection can however provide accurate practical models for any incidence direction with only moderate effort[1,2]. The principle is to establish a mathematical pattern with which these distribution functions evolve, as incidence direction changes rather, than aim for a single universal function. This will be demonstrated with examples .

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In terms of their scattering mechanisms there are a wide variety of diffuse systems, found in nature and manufactured, but those of interest in transmitting and spreading light with polymers tend to have some common features, which simplify the characterisation process. We focus on this issue in the next section where we also address the question of *what is an ideal diffuser material for a skylight and a lamp or luminaire cover*? The study of oblique incidence has some ramifications also for how the BRTF (or more particularly the reference frame for solid angles) is defined, which is examined. We became aware of this issue when calibrating our photogoniometer for oblique incidence, along with analysing the energy and lumen dispersion into the forward hemisphere.

Insights into a relatively new class of translucent materials which better approach the ideal, and their comparison to more traditional pigmented diffusers, is the central aim of this paper. These utilize polymer particles which are themselves clear and have refractive indices quite close to that of the host polymer. Their other attraction is they can survive temperatures which melt the host polymer and thus can be added to extruded and injection moulded sheet in a very simple manner, by adding to the original moulding beads when they are formulated. The special new class of materials on which this aspect of the study were based, were supplied by Roehm –Hass[3] and are now available under the name Plexiglass.

2. DEFINING THE IDEAL LIGHT DIFFUSER MATERIAL

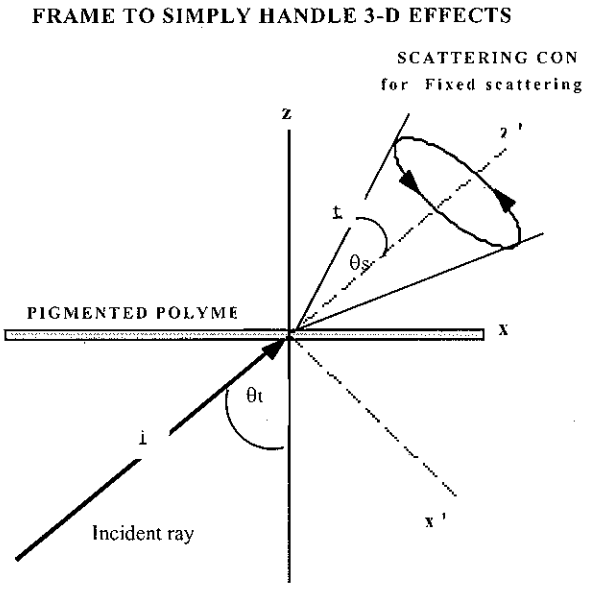


Figure 1 Frame of reference for data modelling and measurements.

The basic measurement geometry is defined in figure 1 where we refer measured transmitted fluxes (Lumens or Watts) on the detector, to a scattering angle θ_S. This deviation angle plus an azimuthal angle ω about the beam direction, provide spherical polar coordinates in the frame defined by x', y', z'

with z' the beam direction and $y' = y$. Use of this cone does not imply symmetry about the incident direction (then $T(\theta_S, \omega)$ is independent of ω), but such symmetry for uniformly pigmented sheets is commonly found in practice, except for high angles of incidence for reasons we touch on later. It is used here since it applies in this work.

As well as a full transmitted distribution it is also useful to have a simple comparison between materials via a single parameter related to this angular spreading. For this purpose a "half angle" $\theta_{1/2}$ which is defined and measured for normal incidence, is introduced. It is defined as the angle θ_S at which measured intensity on a detector is at 50% of the peak value (in an axially symmetric transmitter). A fully diffuse, Lambertian diffuser would have $\theta_{1/2} = 60°$, but typical skylight and luminaire polymers have a half angle well under 60°. This is simple for experimental purposes as it is only necessary to scan a detector along any arc with the sample illumination point at its centre, until the half intensity θ_S is reached. If a material clearly does not scatter symmetrically about the normal for normal incidence, as occurs in some diffusers, we suggest $\theta_{1/2}$ be replaced by the mean of its value obtained by scanning in two perpendicular planes. Another type of half angle is in fact more helpful but less direct, as it requires an integral and hence data over a wide range of angles. This is the cone angle inside which is 50% of the transmitted Lumens (or Watts). Note that both of these half angles will change a lot as angle of incidence changes. Another parameter called "diffusion factor" has been used in the past to quantify transmittance isotropy. It is defined by the average of luminance measured at 20° and 70°, divided by the luminance at 5° for normal incidence. We find in practice this parameter is not any more easy to acquire, and is not as easily generalised to arbitrary incidence angle since it is locked into a relativity to the surface normal which is not appropriate in practical diffusers where the peak follows the beam direction. Most importantly it may cause ambiguities and does not discriminate well among many diffusers we have dealt with even at normal incidence. This is because the 70° luminance value is negligible in many samples, even when there are large variations in the "half angle" $\theta_{1/2}$. Thus for a simple and useful experimental comparison of quality we recommend that just two parameters be obtained, both for normal incidence (i) photopic hemispherical transmittance, τ_{HP} (ii) the half angle, $\theta_{1/2}$. τ_{HP} is the integrated hemispherical spectral transmittance weighted by the photopic sensitivity of the human eye but can be measured directly with special detectors. Two additional parameters can give a complete quality picture. Firstly, Ts the specular component of transmittance, may be added if reliably measured. Ts is often small in the pigmented polymer sheets used in lighting and some measurement techniques may include non specular light causing errors (see below). Secondly CIE colour co-ordinates of transmitted light or equivalent should be given, with a close to neutral "white light" value represented by $x = 0.33$, $y = 0.33$, $z = 0.33$, being the general preference. Spectrally, this requires a flat plot of hemispherical transmittance versus wavelength in the visible range, if daylight is the incident source.

An ideal light diffusing material should transmit no visual detail or a weak residual image of the source and the source characteristics, such as a lamp filament or the very bright luminance zone of the sun. It should spread the light uniformly or as desired into a space with no sharp peaks due to a specular term. A bonus for both illumination and visual appeal, tailored to the space boundary details and the luminaire or skylight position in the room, would be reasonable control over the half angle through simple material modifications. From an energy efficiency viewpoint the photopic hemispherical transmittance (τ_{HP}) which gives the fraction of incident lumens transmitted, should be maximised and it is also good if the incident light CIE colour coordinates are not affected on transmission. An attempt has been made recently to introduce a single optimization parameter[4]. It has maximum value near but not necessarily equal to 1, although it is 1 for a Lambertian diffuser. It is the product of (τ_{HP} - Ts) and a number representing the degree of diffuseness. Unfortunately this second parameter is not easily accessible experimentally. It relates to average path length in the polymer, of transmitted light, and thus links indirectly to $\theta_{1/2}$. A simple experimental optimization parameter for maximising diffuseness and throughput is suggested using a Lambertian diffuser as a reference. Based on consideration and analysis of a few mathematical possibilities it was decided to use H, given as

$$H = \tau_{HP}\left(\frac{\theta_{1/2}}{60}\right) \qquad (1)$$

While H may in principle exceed 1, it is in practice usually well below 1 and caution is advised for general use since a different optimum parameter for lighting in a given situation may be needed. This depends on height and spacing of sources and location relative to walls. For instance 30° may be a better design reference. It is easy to adapt equation (1), which has value τ_{HP} for a Lambertian diffuser, to an optimum lower half angle than 60°. Why not include Ts in this parameter? While Ts can be found experimentally with integrating spheres the measured value can be quite dependent on integrating sphere geometry. That is there is typically a range of low scattering angle dispersed light which contributes to such "specular" measurements. An accurate result for Ts can be found by measuring at a very large distance from the sample. We do this, but it may not be generally practical. Thus for general use and simplicity, total hemispherical transmittance is specified in equation (1). Near "ideal" diffusers would have H around 0.9, or a little higher if antireflection was also employed, but in practice all polymer samples here are well below this. The new diffusers examined in this paper can achieve H up to 0.3, while the majority of traditional diffusers have H < 0.1. A 30° reference angle would double these and may be better for these applications. A clear smooth sheet and a dark sheet each have H close to zero, one for letting through little light, the other for not spreading transmitted light.

The problem to date is that traditional diffusers have a conflict in these twin aims, since if they have high τ_{HP} then they usually have low half angle. The reason is that the traditional pigments such as titanium oxide, barium sulphate or zinc sulphide, added to poly-carbonate or acrylic polymers, increasingly backscatter the light as their concentration is increased, so any attempt to spread the transmitted light further through increased concentration, also decreases the total throughput of light. The poor light output ratio (LOR) of many luminaires is thus in part due to the high diffuseness which is required for glare reduction. These new materials also show a very flat spectral response at visible wavelengths, as desired.

3. THE SAMPLES AND PROPERTY MEASUREMENTS

To exemplify traditional systems and how they contrast with the special properties of the new materials, we measured various $BaSO_4$ pigmented polycarbonate samples. This extruded material is widely used. in skylights and other overhead diffusing applications, but is commonly in the form of multi-wall cellular panels. Various sheet thicknesses from 0.15 mm to 1 mm, and four $BaSO_4$ dopant levels have been studied for a range of angles of incidence.

The polymer doping is achieved with clear polymer particles whose refractive index is close to that of the host polymer, which is PMMA here. The dopant particles are spherical and have much larger diameters (typically 20 to 35 μm) than traditional pigments. They are cross-linked during preparation, which is crucial, since it means most of the polymer spheres can survive typical extruding and injection moulding temperatures of up to 300° to 350° C. Some evidence of particle fragments is found in the moulding beads and final samples, but they do not alter the overall characteristics. These large spheres mean the physical mechanism of dispersion is different to the multiple scattering encountered by the much smaller inorganic particles discussed by Vargas[5]. A full treatment covering multiple successive interactions with these larger clear spheres will appear elsewhere, but it is easy to see by simple mathematical ray analysis, and it can also be observed by eye in a good optical microscope, that most rays undergo very small deviation on passing througb one such sphere. For instance if the index difference to the host is around 0.02, most rays deviate by less than 2° at one sphere, while the few right near the sphere edge may deviate by 4° to 6°. There is negligible backscattering and this underpins their higher H values, since it means hemispherical transmittance τ_{HP} can be very high. τ_{HP} values are over 0.25, but most remarkable is the dependence of τ_{HP} on thickness. One sample below, with constant level of dopant particles was studied at 4 thicknesses; 1.17 mm, 2.02 mm, 3.04 mm and 3.97 mm, and τ_{HP} did not change with thickness within experimental error (± 0.005 in τ_{HP}) . In addition τ_{HP} in this particular system was around 1.2 % higher than the transmittance of a clear sheet of the same host polymer! This is at first puzzling, but may be explained as due to the high but forward half angle reducing exit face reflectance with some even totally internally reflected also ultimately adding to transmitted light. A detailed theory including this feature is under development.

BRTF measurements for various incidence angles are performed using our photogoniometer in narrow beam mode, which has been described in detail elsewhere[1,2] with related principles elucidated by Apian-Bennewitz[6]. To convert the measured intensity on the detector area A_{det} to radiance or luminance we can divide by the solid angle $A_{det}/r^2 = 1/F$ where r is the radius of the scan arc measured from the illumination point. In practice this factor F is always close to the geometric value, but can vary a little due to finite size of the beam and detector so we calibrate F for each incidence direction using τ_{HP} , as measured on an integrating sphere. This process, involving the integration over solid angles in equation (2) for normal incidence and conically symmetric dispersion, gives other insights, especially at higher incidence angles. $T(\theta_S)$ is the signal on the detector at angle θ_S to the beam, divided by the signal on the detector of the direct beam with no sample.

$$\tau_{HP}(0) = 2\pi F \int_0^{\pi/2} T(\theta_S)\sin\theta_S d\theta_S \qquad \text{with} \qquad F \approx \left(\frac{r^2}{A_{\mathrm{det}}}\right) \qquad (2)$$

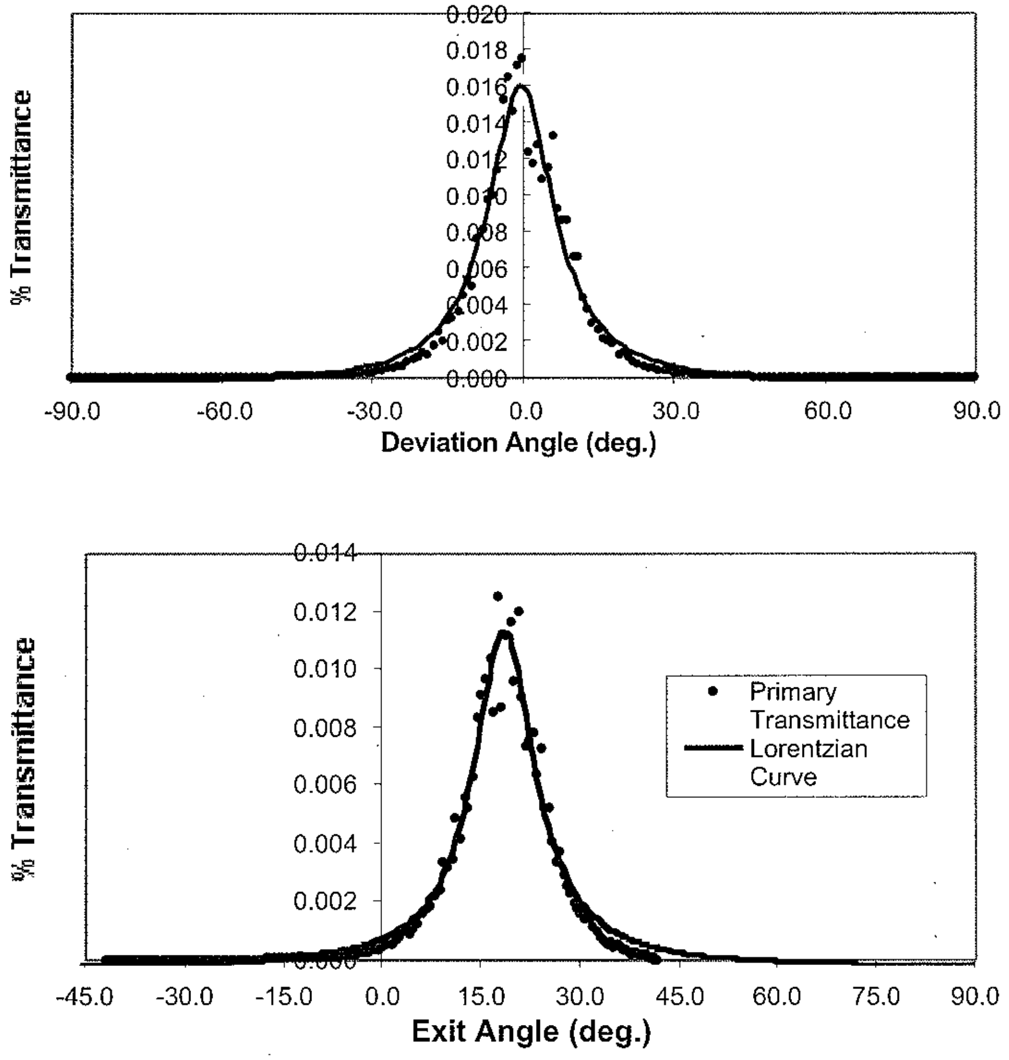
The number found for F is used to determine luminance and BRTF and should be close to the number given by the approximate expression for F. For other angles of incidence the integral is split into three parts according to the cone angle θ_S, to allow for parts of the cone intercepting the sample surface for higher θ_S. As a result the azimuthal integral given by $\int d\omega \neq 2\pi$ as in equation (2) for two of these integrals, but depends on θ_S and θ_i. Note that we always reference measurements and solid angles to the incident beam direction or more precisely the direction in which transmittance peaks. Traditionally it is usual to reference solid angles for BRTF to the sample normal. This is a major issue in its own right. It can lead to problems at oblique incidence if care is not taken. Calibrating against integrating sphere results using the appropriate modifications of equation (2) gave badly incorrect F values unless this frame of reference is used, and it is this that alerted us to the issue. Two frames of reference may be needed sometimes, with the second to allow for a diffuse component centred around the normal if it exists. In practice we rarely find this for commercial polymer diffusers for diffuse transmittance, but do for diffuse reflectance.

4. RESULTS AND DISCUSSION

Figures 2a and 2b show the scattering profile $T(\theta)$ for a sample of polymer doped with polymer spheres for normal and 30° incidence. They have been fitted with the Lorentzian of equation (3) which we

$$T(\theta_S,\theta_i) = T_0 + \frac{2A(\theta_i)}{\pi}\frac{w(\theta_i)}{4(\theta_S-\theta_i)^2 + w(\theta_i)^2} \qquad (3)$$

Figure 2 Fit of equation (3) to data for a) normal incidence b) 30° incidence



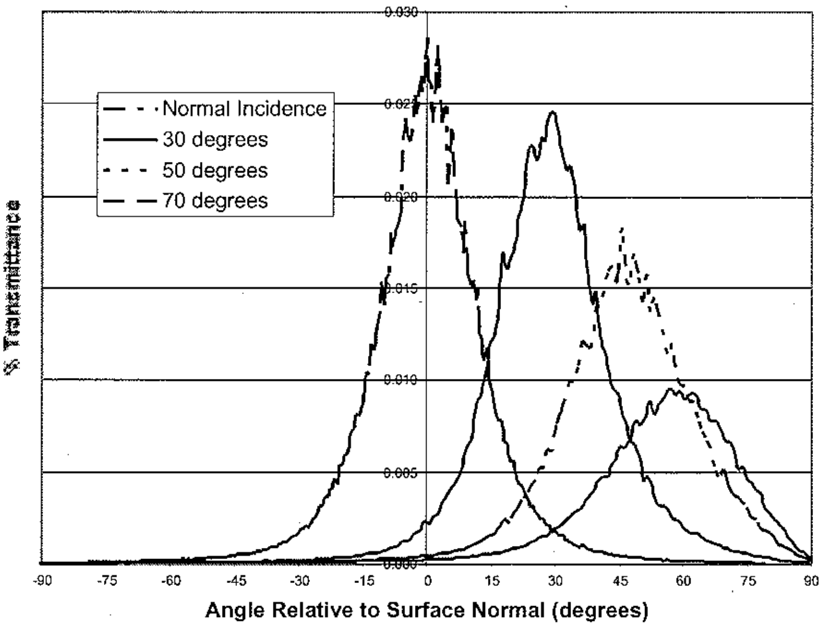


Figure 3 Measured intensity vs scattering angle for four angles of incidence in PMMA doped with clear spheres.

find works well for the polymer and the inorganic doped systems when it is referenced to the beam direction defined by θ_i. This equation also allows development of a simple scheme for any angles of incidence. Rather than attempt to obtain a universal BRTF equation which has serious fundamental problems as noted above, for angle of incidence changes, empirical relations are developed for the parameters A and w in equation (3) as a function of θ_i [or (θ_i,ϕ_i)] . It is then very easy to carry out simulations, with three [four] equations together effectively defining all required data. This also avoids the need for cumbersome data matrices. A detailed study on this issue is forthcoming. Figure 3 shows data as angle of incidence changes in a sample of PMMA doped with cross linked large spheres. These curves have also been accurately fitted with equation (3).

Table 1(a) shows for normal incidence the two key parameters, hemispherical transmittance and half angle, given by $w(\theta_i)/2$, using equation (3) in the Lorentzian fits. Examples appear for a polycarbonate sheet near 1 mm thick for 4 different doping levels of $BaSO_4$ decreasing in concentration from sample a to sample d, and for three different doping levels of Plexiglas sheet, one at four thicknesses. H the joint normal incidence optimization parameter of equation (1), is also given. The polymer doped with large polymer spheres shows a number of important differences, specially the ability to combine large values of both key parameters at appropriate doping levels and hence achieve higher H. The major contrast is in the weak effect of both doping level and thickness on τ_{HP}, and the ability to vary half angle significantly with minimal change in τ_{HP} for the sphere doping, whereas in the traditional systems τ_{HP} falls off strongly as $\theta_{1/2}$ increases. The largest half angle and H value is for the more heavily doped Plexiglas at normal incidence. Table 1(b) shows results for a 3.9 mm thick PMMA sample doped with spheres for three angles of incidence. At 60° there is a fall off in τ_{DH} which is due to more than just change in direct path length plus the extra front surface reflection at 60°. Thicker poly-carbonate is used in practice in a single layer, but the layer here is used in multi-wall sheets. H does not improve in thicker samples. Note that half angle rises more rapidly as angle of incidence increases.

Table 1(a) Comparison of basic performance parameters for diffuse polycarbonate with $BaSO_4$, and PMMA doped with polymer spheres. Small letters denote different concentrations of the pigment or spheres. One of the PMMA set is shown for 4 thicknesses.

Sample	**Hemispherical Transmittance τ_{DH}**	**$w = 2\theta_{1/2}$ degrees**	**Joint performance H(eqn 1)**
1 mm PC sample a	0.759	18.7	0.12
b	0.765	13.3	0.09
c	0.833	6.4	0.04
d	0.874	2.6	0.02
PMMA + spheres a	0.915	21.9	0.16
b	0.884	35.8	0.26
c 3.97 mm	0.939	16.2	0.13
c 3.04 mm	0.937	11.6	0.09
c 2.02 mm	0.947	6.8	0.05
c 1.17 mm	0.941	2.7	0.02

Table 1(b) Example of angle of incidence impact on performance parameters

Sample	**Angle of Incidence degrees**	**Hemispherical Transmittance τ_{DH}**	**$w = 2\theta_{1/2}$ degrees**
PMMA + spheres 3.97 mm thick , c as in table 1(a)	0	0.939	16.2
	30	0.863	19.1
	60	0.498	27.7

5. CONCLUSION

A new class of light diffusing materials has been studied based on clear polymer doped with large clear cross-linked polymer spheres and has been characterised for normal incidence and oblique incidence. It outperforms traditionally pigmented poly-carbonate in its ability to maintain high hemispherical transmittance while varying the spread of transmitted light. A new simple approach to modelling pigmented polymers for lighting has been outlined. Other types of BRTF profile are found in some glazing systems, for example laminted glass diffusers[7]. Due to their lack of any significant diffuse component with a peak normal to the surface a new approach to the BRTF function is suggested. Two parameters have been identified for ease of practical comparison between light diffusing polymers and a way of combining them for overall quality is also suggested. The new polymers allow fewer lamps for specified lux levels with low glare, and hence large energy savings. Smaller skylights can provide the same or higher daylight illuminance with more uniformity and less time of day and seasonal dependence.

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