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Simulation of the thickness dependence of the optical properties of suspended particle devices

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ABSTRACT

Suspended particle devices (SPDs) are able to rapidly switch from a dark bluish-black state to a clear greyish appearance when an AC electric field is applied. Two-flux and four-flux models were used to derive refractive indices and extinction coefficients, as well as scattering and absorption coefficients, of the particle-containing active layer. These entities were used in model calculations to predict direct, total and diffuse components of transmittance and reflectance, along with color appearance and haze, as a function of the thickness of the active layer. An optimum thickness for optical contrast of the SPD was determined in this way and was found to be in the range of 200–300 nm. The devices exhibit significant haze particularly in reflection.

1. Introduction

Green nanotechnologies have a number of overarching goals and can contribute to the preservation of the world's ecosystem by reducing the use of fossil fuels, enhance environmental sustainability, and improve life and wellness for humanity. Specifically, optical materials are desired for energy efficient smart windows, which are able to provide good visual indoors–outdoors contact and daylighting [1]. About 40% of the primary energy use in the European Union is connected with the buildings sector, and smart windows are an innovative fenestration technology that is able to reduce energy use, and hence carbon dioxide emissions, due to lighting, heating, and air conditioning. At the same time, indoor comfort levels can be improved by changing the optical appearance of the windows depending on the outdoor temperature and lighting conditions. For example, the need for space cooling and air conditioning during summer months can be reduced with a heavy tint of the window [2].

Three main electrically controlled optical switching technologies are currently researched for smart windows applications. These technologies can change the windows' optical appearance from a clear, bleached or transparent appearance to a colored, dark or translucent state by means of different operation principles. The technologies make use of electrochromic (EC) materials, polymer dispersed liquid crystals (PDLCs), or layers containing suspended particles [3–5]; all of them require transparent conductors as electrical contacts. The suspended particle devices are often referred to as SPDs.

EC devices are able to change their optical properties in a persistent and reversible way due to redox reactions under the action of a small applied voltage. Such devices are used not only for smart windows but also for mirrors and displays. EC devices consist of a multi-layer assembly incorporating solid EC anodic and cathodic films that are joined by an electrolyte layer and typically sandwiched between two panes of glass or polymer. Coloration can be achieved by electrochemical oxidation of an anodic film and electrochemical reduction of a complementary

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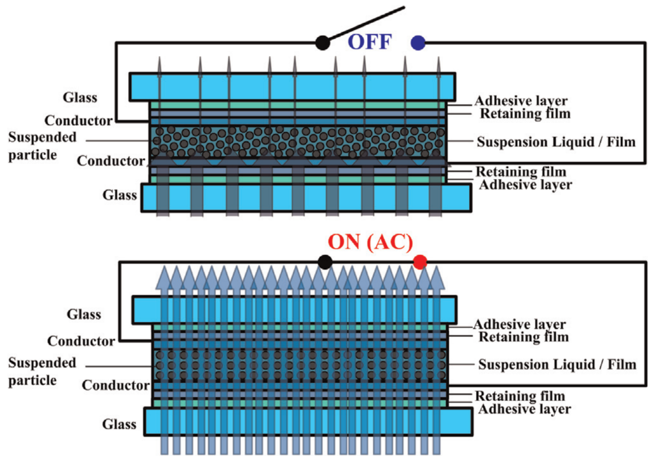

Fig. 1. Schematic multilayer structure of a SPD in "off" and "on" states.

cathodic film and involves transfer of ions into and out of the EC films via the electrolyte layer.

PDLC glazings are comprised of two substrates of glass or plastic joined by a mixture of polymer and liquid crystal (LC) droplets. PDLCs change from a translucent white milky appearance to an almost transparent state when an alternating voltage signal is applied; the LCs are randomly oriented in the former state but aligned under the action of the electric field. The performance of a PDLC-based smart window depends on the size of the LC droplets, which in its turn is related to the curing of the LCs with the polymer [6]. The optical control is mainly confined to visible light, and the solar energy modulation is not large.

SPD technology, which the present study is about, uses the alignment of optically absorbing particles suspended in a cross-linked polymer matrix. Light transmission is controlled by the application of an AC voltage of high amplitude. An SPD consists of 3–5 layers with a schematic construction according to Fig. 1 [7]. The active layer contains needle-shaped dipolar particles of poly-iodides (polyhalide crystals) [8] such as iodoquinine sulfate or herapathite [9,10]. These particles are less than 1 μm in linear size and exhibit large optical anisotropy. The optical anisotropy of herapathite has been studied in detail [11]. Other related compounds are also of interest for SPDs [12,13]. Preferably, the size of the particles should be less than 200 nm in order to minimize light scattering and avoid non-desired haze. The active layer is laminated between two optically transparent and electrically conducting films (such as ITO) and is positioned between two substrates of glass or polymer as shown in Fig. 1. In the "off" state, the suspended particles are randomly oriented and absorb and scatter visible light. The SPD then shows a bluish-black dark color, since most of the light is absorbed by the SPD layer. The scattering effect is mainly due to the particles and is most prominent at short wavelengths. In the "on" state, when the electric field is applied, the particles line up perpendicularly to the substrates and then more light is allowed to pass so that the transmission is increased. The optical properties are prone to be anisotropic when the device is in its activated state. However, for the purposes of this study, the medium can be considered as an effective isotropic material, both in the clear and dark states. There is no memory effect for the optical properties, and hence the electric field must be maintained for keeping the SPD transparent [14].

SPDs switch from dark to bleached or clear state upon the application of a voltage. The much lower transparency in the SPDs' bleached state, compared with the one achievable in ECDs, and undesired haze are the main drawbacks of the SPD technology. Among its advantages, optical response times are around 1–3 s, which is in the same range as for PDLC-based devices and much shorter than for EC devices. The dark bluish-black state, rather than the translucent milky appearance achieved with PDLCs, makes SPD technology suitable for automotive applications. The SPD's size does not affect its switching time to any significant extent, but low temperatures tend to give slow switching.

The light absorbing particles in the SPDs lead to extinction (absorption and scattering) of the incoming electromagnetic radiation. Absorption is a process that removes the radiant energy from the electromagnetic field and transfers it to other forms of energy, whereas scattering redirects the radiation. In previous work, we obtained a detailed set of data on scattering and absorption coefficients for an SPD by inversion of experimental reflectance and transmittance data using two-flux and four-flux models [15]. The present work is a sequel to this earlier study and uses the previously determined scattering and absorption coefficients to investigate the thickness dependence of the active layer in an SPD, including its optical contrast and haze, by numerical computation using the two-flux and four-flux theory.

2. Experiments and calculations

2.1. Optical measurements

Electromagnetic radiation can be specularly reflected (R_{spec}), scattered (S), absorbed (A) or directly transmitted (T_{dir}) when it crosses a medium that is different from the one from which it emerges. These quantities are related by the relation $R_{spec}+A+S+T_{dir}=1$. Materials that are able to control any of these four parameters by means of an external stimulus are known as "chromogenic" [16]. Optical characterization of the materials includes measurements of total and diffuse transmittance (T) and reflectance (R)—denoted T_{tot}, T_{diff}, R_{tot} and R_{diff}—and are usually carried out with an integrating sphere based spectrometer [17]. Direct transmittance and specular reflectance are computed by subtracting the total and diffuse components. The sum of the diffuse transmittance and reflectance is the parameter S related to scattering, i.e., $S=T_{diff}+R_{diff}$. Absorption can be therefore derived from R_{spec}, T_{dir} and S.

The SPD investigated below has an active area of 28×22 cm^2 and a thickness of 300 μm; its details have been reported before [7,15,18,19]. Specifically, the SPD is a CriRegulite device supplied by CRICURSA (Cristales Curvados S.A., Barcelona, Spain), which is a licensee of Research Frontiers, Inc. (Woodbury, NY, USA). The SPD was operated with a sinusoidal signal at 50 Hz and a peak voltage U between 0 and 100 V.

The final goal of the previous work was to decouple the scattering and absorption coefficients of the SPD in both dark ("off") and clear ("on") states [7,15]. In order to accomplish this, we measured T_{tot}, T_{diff}, R_{tot} and R_{diff} in the $300<\lambda<2500$ nm wavelength range for the SPD in its "off" and "on" states by using a Perkin Elmer Lambda 900 double-beam spectrophotometer equipped with an integrating sphere. A Spectralon plate was used as reflectance standard, and corrections for the reflectance of the standard as well as port losses were carried out as previously described [17]. Collimated–collimated (cc) measurements (i.e., recordings of collimated light when illuminating with such light) gave R_{spec} and T_{dir}, and collimated–diffuse (cd) measurements (i.e., recordings of diffuse light when illuminating with collimated light) gave R_{diff} and T_{diff} for the SPD. Luminous (lum) and solar (sol) transmittance and reflectance were obtained by averaging spectral optical data over the spectral sensitivity of the light-adapted human eye [20] and over the AM1.5 solar spectrum corresp-