

## High Transmittance of the Glass Coated by the PMMA Mixed with Silica Gel

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### Abstract

*The transmittance of bare glass was enhanced up to about 20% by coating it with a PMMA (Poly Methyl Meta Acrylate) film mixed with silica gel. This low-refractive-index film greatly reduces total reflection inevitable for bare glass, and thus will be useful for increasing the coupling-out efficiency of OLED.*

### 1. Introduction

A portion of the light emitted by the organic material in OLED is totally reflected during the propagation to the front glass of OLED due to the high-refractive-index materials of the constituent films. Consequently, the coupling-out efficiency of OLED is a mere 15%, approximately. An attempt to increase this efficiency by fabricating micro-sized lenses on the glass to focus the incident light has been reported [1]. But this technique involves many complicated manufacturing processes, and thus is not practical. Another experiment reduced the total reflection during the propagation of light from the oxide layer to the glass by sandwiching a low-refractive-index aerogel film between the oxide layer and the glass [2]. However, this technique involves the supercritical drying process which needs to be performed at a very high pressure of about 70 atmospheric pressure [3, 4]. Furthermore, it also has the disadvantage of the brittleness of aerogel. In our experiment, we used PMMA [5] to form a low-refractive-index film. PMMA has a reasonably low refractive index of 1.49 at the wavelength of 656nm (red Fraunhofer wavelength), and a PMMA film can be easily formed on glass by the spin-coating method. We could also achieve a lower refractive index of 1.48 by mixing silica gel clusters into the PMMA solution. Transmittance of the glass coated with PMMA mixed with silica gel was enhanced up to

about 20% compared to the transmittance of bare glass.

### 2. Results

The flowchart for preparing the PMMA plus silica gel solution is shown in Figure 1.

Two solutions are prepared for producing silica wet gel, which eventually will be mixed with the PMMA solution. One is the silica solution which contains 10mL of TEOS (Tetraethoxysilane) [6] and 8mL of ethanol. The other solution is the catalyst solution containing 7mL of ethanol, 14mL of water, 0.03mL of 30% aqueous ammonia, and 0.2mL of 0.5M ammonium fluoride. Silica wet gel is formed when the catalyst solution is slowly added (a few drops, a drop every 20 min.) to the silica solution while stirring. The water inside the silica gel can be extracted by washing it with ethanol several times. Afterwards, the solvent in the silica gel is exchanged with n-hexane [6]. This solvent exchange process is necessary for the surface modification of the silica gel by TMCS (Trimethylchlorosilane) [6] solution. In the surface modification process, hydroxides ( $\text{OH}^-$ ) on the surface of the silica gel are removed. These hydroxides combine with the vapor in the air and induce deformation of the silica gel when it is exposed to the air [7, 8], and thus needs to be removed. The solvent exchange with n-hexane before the surface modification process is necessary because TMCS solution does not dissolve in ethanol, but does so in n-hexane. Another solvent exchange process with THF (tetrahydrofuran) [6] solution is followed. Afterwards, the silica gel is redispersed in THF solution using the energy of ultrasound. This solution is then mixed with the PMMA solution made by dissolving PMMA pellets [5] in THF solvent. This mixed solution is slowly dried until it has a suitable viscosity for spin coating.

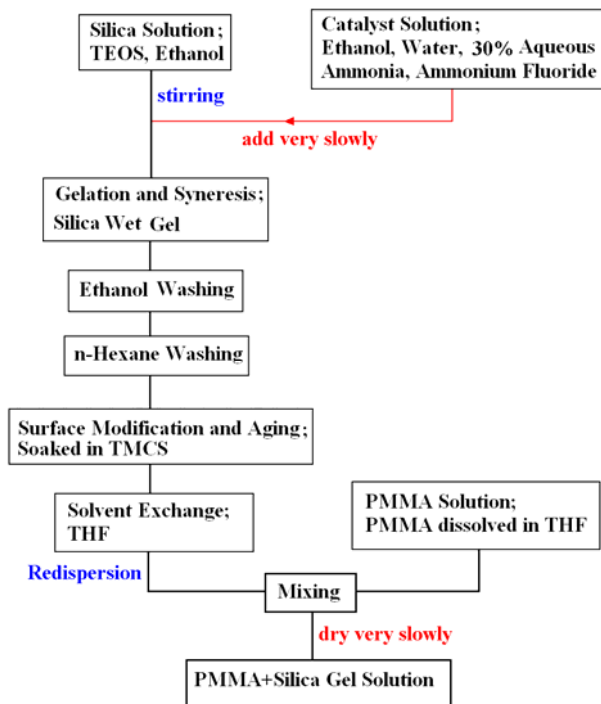


Figure 1. Flowchart for the preparation of the PMMA plus silica gel solution.

The PMMA film is formed on glass substrate by using a spin coater. Film thickness was determined to be about 0.014mm by comparing the weights of the coated glass and glass substrate. The distribution of the silica gel in PMMA film is observed by TEM(Transmission Electron Microscope), which is shown in Figure 2.

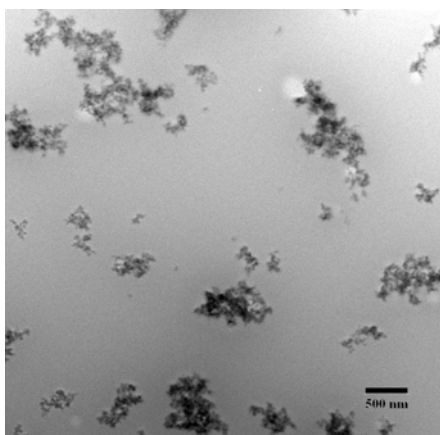


Figure 2. TEM picture of clusters of silica gel in the PMMA film.

Wavelength dependence of the refractive index for PMMA film mixed with silica gel is shown in Figure 3. The refractive index was measured with an ellipsometer (Model MG1000 of Nanoview). In this figure, the refractive index of PMMA film without silica gel is also shown for reference. The refractive index of PMMA film at the red Fraunhofer wavelength (656nm) is about 1.49, which is lower than the 1.52 of bare glass. We can see that mixing the PMMA film with silica gel further lowers the refractive index to 1.48.

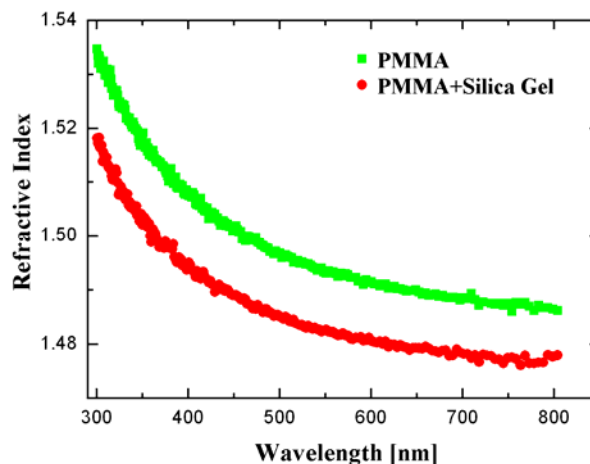


Figure 3. Wavelength dependence of the refractive index for PMMA and PMMA+silica gel films.

The transmittances of laser light (He-Ne laser) through bare and coated glasses were observed by using an experimental set-up as shown in Figure 4.

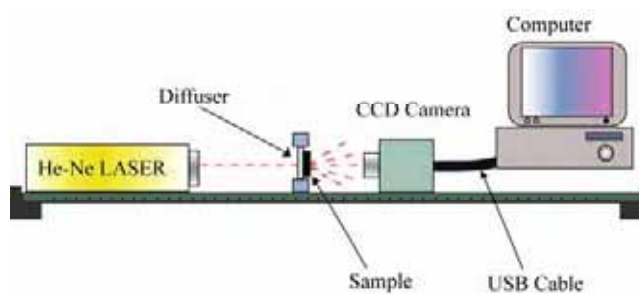
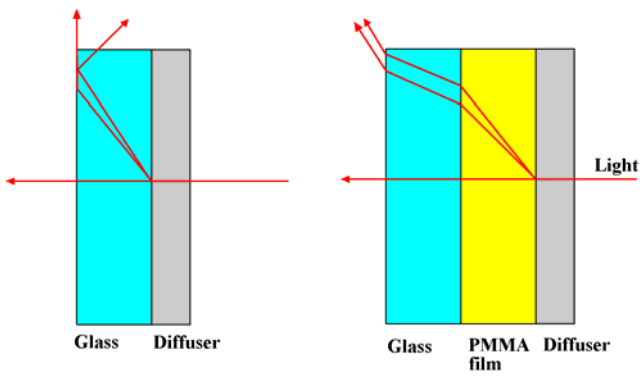


Figure 4. Experimental set-up for measuring the transmittance of laser light through bare or coated glass.

The diffuser scatters the laser light, which enters the bare or coated glass in all directions; this simulates OLED, in which point source light generated at the organic material scatters and enters the neighboring layer of film [9]. Some of the incident light coming into contact with bare or coated glass is totally reflected, and consequently cannot be transmitted to the air. Difference in total reflection for bare and coated glasses is schematically shown in Figure 5. We can see that some of the light totally reflected in bare glass can be converged by the coated film and transmitted to the air in coated glass.



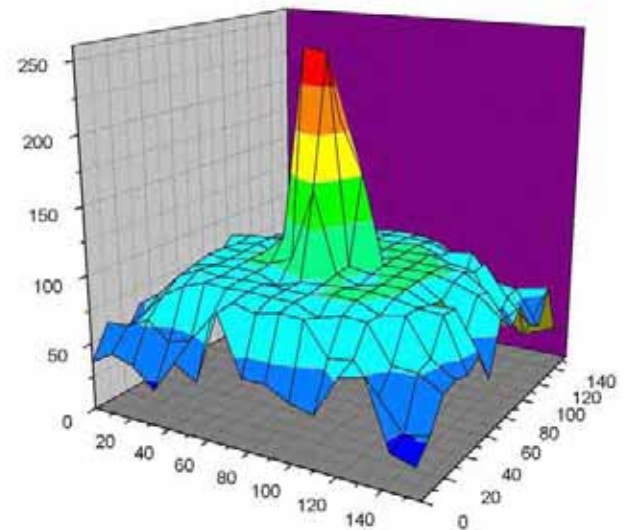
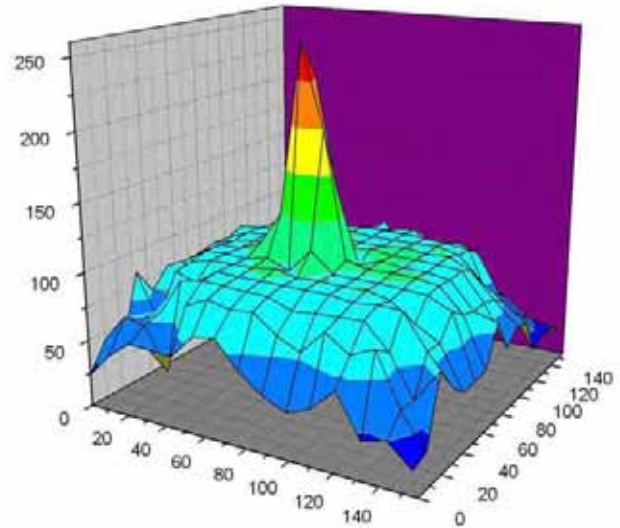
**Figure 5. Dispersion of the light source through bare glass (left) and glass coated with the PMMA mixed with silica gel (right).**

Intensity distributions of the transmitted light through bare and coated glasses are shown in Figure 6. Light with an incident angle higher than the critical angle for bare glass can be transmitted through coated glass as shown in Figure 5. Thus, as shown in Figure 6, width in the intensity profile for coated glass becomes much greater than that for bare glass. We can estimate from the intensity profile in Figure 6 that the amount of transmitted light through coated glass is increased up to about 20% compared to that through bare glass.

### 3. Conclusion

OLED has metal, organic material, ITO(Indium Tin Oxide), SiO<sub>2</sub> films, and front glass in this sequence when seen from the bottom. Refractive indices of organic material, ITO, and front glass are 1.7, 1.8-2.1, and 1.52, respectively. 51% of the light produced at the organic material leaks out as a waveguide through the contact space between the

organic material and ITO. Furthermore, 31% of the light transmitted at the front glass also leaks out to the sides of the glass due to total reflection. Consequently, about 15% of the light generated at the organic material is transmitted to the air for display to our eyes. The major reason for this low coupling-out efficiency of OLED is its structure in which light passes through a high-refractive-index material (ITO) to a low-refractive-index material (glass).



**Figure 6. Transmittances of light through bare (top) and coated (bottom) glasses.**

In our experiment, we estimated the reduction in total reflection benefiting from the insertion of a low-refractive-index film between the ITO and the glass in OLED. The ideal material for this inserted film is silica aerogel [2] whose refractive index is in between 1.01 and 1.1. However, to form this film, we need the supercritical drying process which needs to be performed at a very high pressure of about 70 atmospheric pressure [3]. Thus, we used a polymer material, PMMA, a thin film of which can be easily produced by the spin coating method. In addition, we tried to form a film with a lower refractive index by mixing silica gel clusters into the PMMA solution. By coating the glass with PMMA mixed with silica gel, we could achieve an enhanced transmittance of light up to about 20% compared to that through bare glass. We could conclude that the coated film with low refractive index greatly reduces the total reflection. PMMA coating is easily performed by the spin coating method, and gives a relatively high transmittance compared to other complicated techniques [1, 2]. Our film structure of low refractive index will be useful for enhancing the coupling-out efficiency of OLED.

#### 4. Acknowledgements

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