Design of ZnO multilayers with different porosities for UV absorbing transparent glasses

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Abstract- **We present ZnO multilayers with different porosities on glass substrate for high efficient UV absorbing transparent glasses. Diffraction efficiencies of the proposed ZnO multilayers were calculated using a rigorous coupled wave analysis method. The results show the effect of design parameters such as effective index, porosity and thickness on the optical characteristics. The analysis based on volume averaging theory also supports the calculation results. Detailed design guidelines for optimum geometry are also discussed.**

I. Introduction

Harmfulness of ultraviolet (UV) rays, which causes deterioration of physical/chemical properties in several materials and health problems on the human body, has been already noted. Absorbing materials (e.g., zinc oxide (ZnO), cerium oxide (CeO_2), titanium dioxide (TiO_2) and so on.) for UV shielding show low transmittance in the visible ranges due to high refractive index. For transparent UV absorbing applications, density control of UV absorbing materials is crucial to obtain both strong absorption and high transmission in the UV and visible region, respectively.

A simple method for controlling density is oblique angle deposition (OAD). The OAD technique has the ability to generate porous nanostructures, caused by the self-shadowing nature of the deposition process. Furthermore, the density can be controlled by changing the porosity with the angle of incoming particle flux [1, 2].

Prior to apply OAD in transparent UV shielding, the optical design of such structures for improving optical properties needs to be optimized with important details related to the transmittance/absorbance. In this paper, we numerically designed optimum geometry of ZnO multilayers with different porosities for UV absorbing transparent glasses.

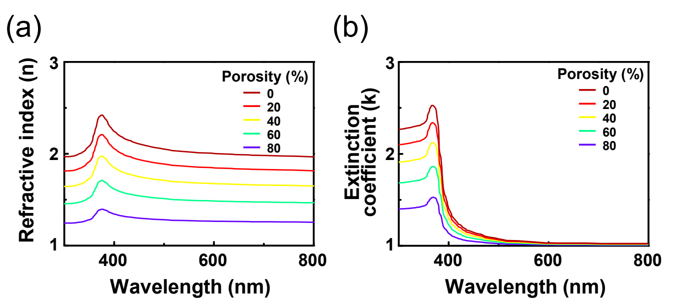

Fig. 1. (a) Calculated refractive indices (RI) and (b) extinction coefficients of ZnO with different porosities (PR) as a function of wavelength.

II. Simulation Result and Discussion

Fig. 1 shows calculated refractive indices (RI) and extinction coefficients of ZnO with different porosities (PR). The volume averaging theory (VAT) was used to calculate effective complex index (n, k) of ZnO with the porosity [3, 4]. The result that the higher PR has the lower n, k is obtained.

Fig. 2(a) displays RI profiles of a conventional ZnO (PR 0%) thin film (dash line) and proposed ZnO multi-layers (solid line) on a glass substrate. UV absorbing oxide materials including ZnO has a refractive index of ~2.1, which is much higher value than that of glass substrate (n_g ~1.5), resulting in strong Fresnel reflection. Such negative effect can be minimized by introducing ZnO multi-layers with PR of 60, 80% and a thickness of 300 nm. Due to the fact that PR 60% and 80% of ZnO provide the effective RI of ~1.51 and ~1.28, the difference of RI between the UV absorbing layer and the glass substrate is reduced by gradual RI change. To calculate diffraction efficiency of ZnO multilayers on glass substrates, the rigorous coupled-wave analysis (RCWA) method was employed. For comparison, three transmittance spectra of glasses with PR 0%, PR 60% ZnO thin-films and PR 60 / 80% ZnO multilayers were calculated, as depicted in Fig. 2(b). Although mostly perfect UV absorption was achieved in the PR 0% ZnO thin film, the poor transmittance in the visible range is obtained as we expected. On the other hand, by using the ZnO multi-layers, the transmittance in the visible region is improved with maintaining UV transmission of less than ~8%. By using only PR 60% ZnO thin film, the transmittance in the visible region was also improved, however it has lower than that of PR 60 / 80% ZnO multilayers.

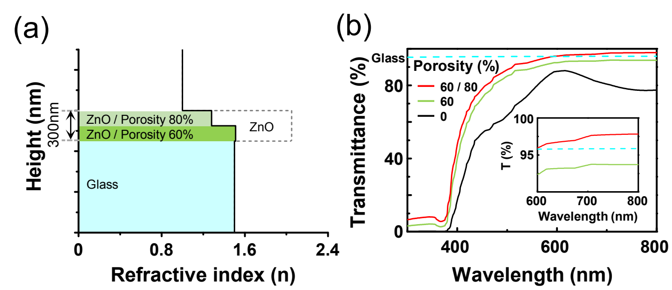

Fig. 2. (a) Refractive index profiles versus height UV absorbing transparent glass with a conventional ZnO thin film (dash line) and ZnO multilayers with different porosities (solid line). RI of each material at 500 nm were considered. (b) Transmittance spectra of glasses with the PR 0%, PR 60% ZnO thin-films and PR 60 / 80% ZnO multilayers. The thicknesses of thin films and multi-layers are 300 nm.

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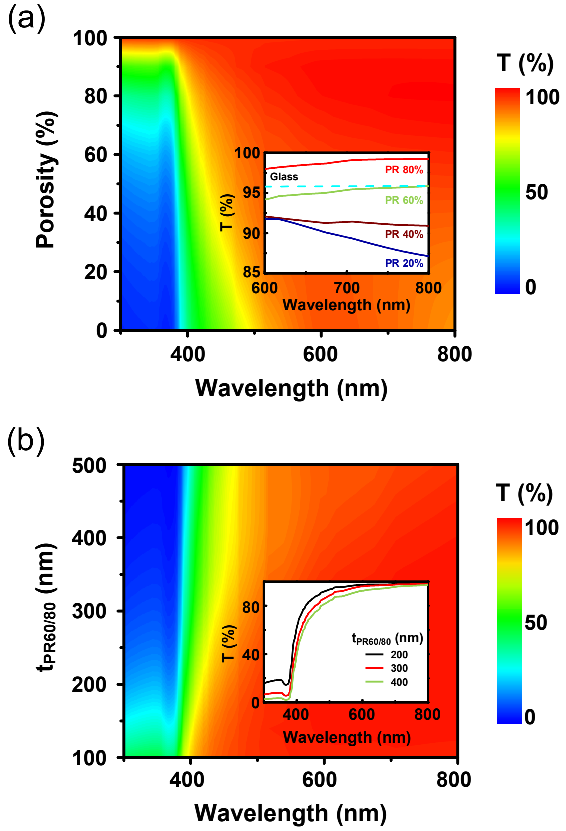

Fig. 3. (a) Contour plot of variation of transmittance of PR different ZnO thin film with a thickness of 150 nm. Inset shows enlarged transmittance spectra at 600-800 nm. (b) Contour plot of variation of transmittance of PR 60 / 80% ZnO multilayers with different thickness ($t_{PR60/80}$). The thicknesses of each layer of multi-layers are equal.

In order to optimize the geometry of ZnO multilayers, we conducted calculations with different PR and layer thicknesses. The PR has a trade-off between UV absorption and transmission at visible wavelength, due to the change of n, k depending on the PR. The UV protection effect is given by high extinction coefficient of ZnO at UV region, however, the presence of extinction coefficient at 400-500 nm generates an adverse effect on the transmittance in this region. Due to the reason as mentioned above, UV absorption is increased as the PR decreases, while absorption in wavelengths of 400-500 nm is also increased (Fig. 3a). The layer thickness also affects not only the absorption at UV region but also transmission at 400-500 nm since absorption is exponentially proportional to the optical thickness (Fig. 3b). From the calculation results, we choose the optimum geometry with PR 60 / 80% and a thickness of 300 nm.

Fig. 4(a) demonstrates HSV (Hue, Saturation and Value) color coordinate containing chromatic information of PR 60 / 80%, PR 60% and PR 0%. Chromatically, transparency can be evaluated as colorlessness. The closer point to pure white (0, 0, 100) in HSV color map means the more colorless. Compared with PR 0% and PR 60%, the coordinate of PR 60 / 80% is closer to the pure white than PR 0% and PR 60%.

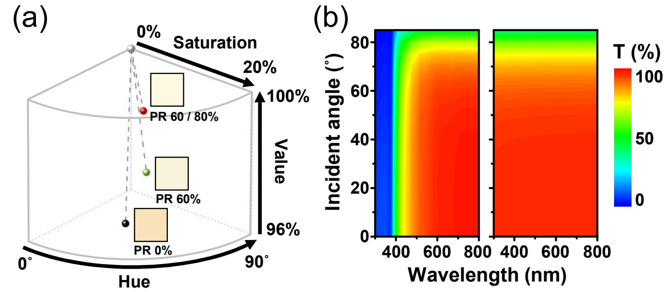

Fig. 4. (a) HSV (Hue, Saturation and Value) color coordinate containing chromatic information and color representation of PR 60 / 80%, PR 60% and PR 0%. (b) Contour maps of angle dependent transmittance of a ZnO HNS arrays with an optimized geometry (i.e., PR 60 / 80% and 300 nm layer thickness) on a glass substrate and bare glass (as a reference).

The influence of the incident angle of light on the transmittance is crucial for the transparent glass applications. The ZnO multi-layers with an optimized geometry (i.e., PR of 60% and 80%, thickness of 300 nm) shows omni-directional high transmittance in the visible region, which is comparable to that of bare glass, while sustaining UV absorption, as shown in Fig. 4(b).

III. Conclusion

In conclusion, we investigated the effects of geometrical parameters (i.e., PR and thickness) of ZnO multilayers on the optical performance of UV absorptive transparent glasses by calculations of effective index and diffraction efficiency using VAT and RCWA method, respectively. From the calculation results, the optimum geometry of ZnO multilayers with 60% and 80% of PR, and a thickness of 300 nm was founded. The concept of ZnO multilayers with different porosities can be used to various UV shielding applications, and our design guidelines would also be helpful to apply OAD in transparent UV absorbing.

Acknowledgment

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