Abstract— Growth of multilayer thin film structures containing dielectric and metal layers using physical vapor deposition is investigated for use in applications requiring the control of thermal and solar radiation propagating through glass windows. In particular, metal-dielectric multilayer structures reflecting UV, near-infrared and thermal radiations whilst maintaining a maximum transmission in the visible range are prepared using both an E-Beam and Thermal evaporator and a RF Magnetron sputtering system. Measured transmittance spectra for the developed structures are in agreement with simulation results and demonstrate that with the use of optimum metal-dielectric layer combination it is possible to realize a coated glass transmitting most of the visible light through and reflecting most of the UV, solar and thermal infrared radiations.

Index Terms—Thin films, Low-E glass, metal-dielectric structures, E-Beam evaporation, thermal evaporation, RF magnetron sputtering.

I. Introduction

The development of specialised glass products and especially the energy-efficient window glazings (e.g. low-emissivity and solar control glass) and related technologies have attracted significant research attention worldwide in recent years [1-5]. To provide energy savings through solar radiation control and/or low thermal emissivity (“Low-E”) properties, modern glazing systems usually employ one or more glass panes coated by thin films which are deposited onto flat glass surfaces. These coatings are custom-designed as optimized sequences of several different optical material layers that (once combined together) will be able to act as mirrors for the infrared wavelengths (thus working as a “heat mirror” [1]), or provide other advanced functionalities (“smart” or electrochromic windows [2, 3]). Glazing systems capable of both solar infrared and thermal (far infrared) radiation control are of particular practical interest due to providing maximized cooling- and heating-related electricity savings in buildings, across a wide range of geographic locations and climates.

In the recent years, many studies have been focused on the deposition of metal-dielectric thin films suitable for use as heat mirrors. Many investigations on metal-dielectric multilayer film structures have been performed during the last two decades [6, 7], which is due to the unique combination of the optical and thermal properties of metal. Several material systems suitable for use in metal-dielectric multilayer coatings, for example, TiO\textsubscript{2}/Ag/TiO\textsubscript{2}, or ZrO\textsubscript{2}/Ag/TiO\textsubscript{2} [6], as well as WO\textsubscript{3}/Ag/WO\textsubscript{3} [1] have been reported.

Titanium dioxide (TiO\textsubscript{2}), silicon dioxide (SiO\textsubscript{2}), silicon nitride (Si\textsubscript{3}N\textsubscript{4}), zinc oxide (ZnO), and aluminium oxide (Al\textsubscript{2}O\textsubscript{3}) are the dielectric materials most often used in glass coatings[8, 9]. The advantages of using oxide materials include a wide range of available refractive indices as well as very high transparency across the visible and near-infrared spectral ranges. One disadvantage of using oxide materials in metal-dielectric systems is related to the risks of oxidizing the ultrathin metal layers placed adjacent to oxide layers during the multilayer deposition. Another disadvantage of placing the oxide layers into contact with metal is the strong tendency of ultrathin metal layers to form nanoparticle islands or agglomerates in the absence of any special “wetting layers” or “seeder layers” which can improve the metal layer morphology[10]. Also, most oxide layers always lose a part of their oxygen content (and thus transparency) when deposited by non-reactive physical vapour deposition processes like e-beam evaporation or sputtering, especially when no oxygen input is used to compensate for oxygen loss.

In this work, we investigate the deposition of ultrathin silver layers in conjunction with non-oxide layers to realise transparent glass that blocks most of the UV, solar and thermal infrared radiations.

II. Simulation and Experimental Setup

A simplified schematic diagram of the developed thin film structure is shown in Fig. 1, illustrating the interaction of the incident sunlight (visible, UV, and IR rays) with a metal-dielectric multilayer system that provides the core functionality in energy-efficient glazings of the solar control type.
Typically, dielectric materials are used in thin-film coatings for their high refractive index, surface thermodynamic properties favourable for silver deposition and good environmental durability [11-13]. However, it is important to note that controlling the thickness of evaporated dielectric layers is always somewhat more difficult compared with other materials. In order to protect the thin film from external environments (moisture, solvents etc.), an adhesive dielectric layer is initially deposited onto the substrate and an oxide dielectric layer is used as the last layer (protector). To attain the desired spectral response for a particular application, different thin layers of optimized thicknesses are sequentially deposited.

The main motivation of this work was to design and demonstrate new material combinations for use in multilayers suitable for making glazing systems with improved optical and thermal properties, namely, achieving more than 80% of visible light transmission, blocking more than 90% of the UV and solar IR radiations, and suppressing more than 95% of the thermal radiations. In addition, the indirect aim was to realise environmentally stable coatings that can be fabricated reproducibly and can compete with current industry-leading energy-saving glazing products. Fig. 2 shows transmission spectra of various thin film coatings developed by the major low-E glass manufacturers in the world. Also shown (dashed line) is the spectral response of an ideal clear energy-efficient glass.

Several metal-dielectric coating structures were simulated and optimized using OptiLayer Software, then the practical structure that has the minimum number of layers was selected and fabricated, using both an E-beam evaporation system and an RF magnetron sputtering system, for use in advanced glazing systems.

Simulated transmittance spectra of several multilayer thin films are shown in Fig. 3. All these thin film structures feature metal (M) layers surrounded by dielectric layers and protected from environmental effects by a top dielectric layer. During the simulations, the properties of single layers of metal on glass substrates were first investigated in order to (i) calibrate the thickness control monitors of the E-beam evaporation system and the RF magnetron sputtering system, and (ii) analyse the behaviour of ultrathin metal layers. It was found that evaporated metal layers behaved consistently with theoretical models for layer thicknesses exceeding 15-18 nm, and that the ultrathin (less than about 20 nm) metal layers typically lost a large part of their reflectivity across the IR spectrum, if deposited onto glass or oxide substrates (TiO₂, Ta₂O₅, Al₂O₃ etc.). This was not unexpected since a number of literature sources have reported similar observations with ultrathin silver layers [2, 8, 9 and 13].
(Glass/D1/Ag/D1/Ag/D1/D2) containing two ultrathin silver layers and four dielectric layers (denoted D1 a dielectric of a high refractive index and D2 is dielectric material of a low refractive index) was selected for the experimental deposition.

It is important to note that different deposition techniques can be used for producing thin films of metals, semiconductors or dielectric materials. However, the most common technique is the physical vapour deposition (PVD), such as thermal evaporation, e-beam evaporation and sputtering [7, 14]. Using our dual thermal/e-beam evaporation system enabled us to evaporate (KVE-ENT 200hl Korea Vacuum Tech), in the same vacuum chamber, up to four different materials through electron beam evaporation and one additional material via thermal evaporation. On the other hand, our RF magnetron sputtering system (KVS-T4065 Korea Vacuum Tech, LTD) has two thickness monitors, namely a quartz monitor and a laser reflectometry based monitor, which enabled the thicknesses of the various deposited layers to be controlled within ±2.0 nm accuracy. Thin films were grown by all three methods (RF sputtering and e-beam/thermal evaporation) on unheated glass substrates, at base vacuum pressures achieved in both deposition chambers being near 1×10⁻⁶ Torr, and the growth rates were typically between 0.5-1.5 Å/s. The argon (Ar) partial pressure used during the sputtering processes was between 1-2 mTorr.

The transmission spectra of the developed thin films measured using Agilent Cary 5000 UV-VIS-NIR and Beckman Coulter DU 640 B UV/Visible spectrophotometer. The simulation software was subsequently used to derive the actual layer thicknesses of both developed films by fitting the measured transmission spectra with simulated transmission spectra of similar layer structures.

It should be noted that the performance of heat mirrors and other edge-filter-type multilayer systems depends strongly on the individual layer thicknesses as well as on the quality of layer interfaces. Therefore, a set of calibration procedures was designed and implemented to measure the tooling factors of the quartz crystal microbalance sensor for every material type used. We found that the tooling factors were strongly dependent on the substrate temperature, and that most of the metal-dielectric systems performed best when deposited at room temperature. Therefore, particular attention was paid to the calibration of all relevant tooling factors for the room-temperature processes. It is also important to notice that the different materials used had different tooling factors even when evaporated using the same e-beam boat location. This was because of the tendency of the e-beam boat location to sublimate and even propagate around shutters. The same problem of calibration was also encountered in the sputtering process.

### III. Results and Discussion

The measured transmission spectra for both developed thin films and the theoretical (simulated) transmission spectrum for the selected design type are shown in Fig. 4. Some discrepancies between the measured transmission and the model in the UV range (below 400 nm) were caused by the fact that the modeling software calculated the power transmission into the “semi-infinite substrate” only, and the UV absorbance of glass itself was not accounted for in these calculations. Also, we multiplied the calculated transmittance at every wavelength by a factor of 0.96, to account for the back-of-substrate reflectivity at normal incidence, after which the experimental results agreed closely with the predictions. The discrepancies between the measured transmission spectrum and the model could therefore be attributed largely to layer thickness errors (both systematic and random), which are almost unavoidable in real deposition experiments. An attempt was made to fit the thickness errors by way of finding the “real” layer thicknesses that would bring experimental results into very close agreement with the model. For this reason, our work is still ongoing on the thickness measurements calibration improvements of evaporated D1 and D2 layers in order to achieve better experimental results.

It is noticed from Fig. 4 that for the thin film developed using the E-beam/thermal evaporator the maximum visible transmission is 84%, which is slightly higher than the visible transmission of the thin film deposited using sputtering (around 80%). This difference in transmission in the visible range was mainly due to the higher risk of source-material cross-contamination in our RF magnetron sputtering chamber compared to the risk of such cross-contamination in the E-Beam/Thermal Evaporator chamber, due to the systems’ design features. However, it is noticed that the latter thin film exhibits a flat visible response and a broader visible bandwidth in addition to better stability and durability, compared to the thin film deposited using E-beam/thermal evaporation. It can also be noticed that both thin films have excellent UV, solar IR and thermal IR isolation properties.

![Fig. 4. Simulated and measured spectral transmittance curves for the optimized multilayer thin films described by formula (Glass/D1/M/D1/M/D2).](image)

### IV. Conclusion

Thin film coatings suitable for controlling thermal infrared and solar radiation have been investigated theoretically and experimentally. Transparent films of heat-mirror type have been designed and fabricated using multilayer metal-dielectric sequences of structure (Glass/D1/M/D1/M/D2). Thin films have been deposited using a combined E-beam, thermal...
evaporator system and sputtering process. The optimization of optical transmission spectra as well as fitting of actual individual layer thicknesses deposited has been carried out using standard thin-film design software packages. The transmission spectra of the developed coatings predicted by theory have been rather close to these measured experimentally. Experimental results have shown that the thin film developed using a dual E-Beam/thermal evaporation system exhibits better transmission (84%) in visible range compared than that of a thin film developed using sputtering (around 80%). However, the RF sputtered thin film is more stable and durable, and exhibits a flat visible response and a broader visible bandwidth, in comparison to the thin film deposited using E-beam/thermal evaporation.

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