Thermochromic Film Optimization by Computational Simulation

Yuki NAOI＊, Jun AMANO＊＊

要旨

建築物や自動車での不必要なエネルギー消費を防ぐには，夏場の暑い時間帯には太陽からの熱線を遮り，寒冷な季節には屋内に透過させるスマートウィンドウが効果的である。特にエネルギー効率の高い，省エネビルの実現のためには，スマートウィンドウが必要である。従来の一般的な窓をエネルギー効率の高いスマートウィンドウに変換するには，サーモクロミック材料を用いた高効率かつ透明なフレキシブルウィンドウフィルムが望まれている。サーモクロミック材料は，外部の温度変化により，追加制御なしでその光学特性を変化させることが可能である。二酸化バナジウム（VO_2）は絶縁体から金属への相転移でシャープな相転移特性を示し，可視光領域における高い透過性を保ちながら，近赤外光領域においては絶縁体から金属への相転移前後で大きな透過率変化を示す。このことから，単結晶二酸化バナジウム（VO_2）ナノワイヤーとポリマーを組み合わせたフレキシブルポリマーコンポジットは，最も有望で高安定性なサーモクロミックウィンドウフィルムと考えられる。本論文では，高エネルギー効率スマートウィンドウに使用する高性能サーモクロミックフィルムの実現のために，VO_2ナノワイヤーの最適形状を探索するためのコンピュータシミュレーションとシミュレーションの結果に要るVO_2ナノワイヤーの最適形状について報告する。

Abstract

In order to reduce unnecessary energy consumption in the buildings and automobiles, smart windows that reflect heat from the sun on hot summer days but let in the heat in colder weather provide a good solution. Especially, to create highly energy–efficient buildings, it is essential to incorporate smart windows. Window films by thermochromic materials with high efficiency, transparency and flexibility are highly desirable to convert non-smart windows to energy efficient smart windows. Thermochromic materials change its optical properties according to the external temperature without any additional input signals. Vanadium oxide (VO_2) particles embedded flexible polymer composite films are most promising thermochromic window films due to a sharp phase transition of insulating to metallic phases with visible transparency and a large change in transmittance at near-infrared (NIR) wavelengths before and after the metal–insulator phase transition. Especially, single crystalline VO_2 nanowires embedded polymer composite films are expected to be highly stable and efficient. This paper describes computational simulation for investigation of the optimum structure of VO_2 nanowires in order to obtain high performance thermochromic polymer composite films for highly energy-efficient smart windows.

＊Industrial Optical System Business Headquarters
＊＊Konica Minolta Laboratory U.S.A., Inc.

1 Introduction

Many buildings have large windows for good daylight penetration and attractive appearance; however, the large window areas often result in poor energy–efficiency of the buildings. Smart windows that reflect heat from the sun on hot summer days but let in the heat in colder weather provide a good solution to create more energy–efficient buildings. Smart windows change light transmission properties in response to external heat, light and electricity. There are several technologies can be used for smart windows utilizing electrochromic, photochromic and thermochromic materials[1]. Thermochromic materials change its optical properties according to the external temperature without any additional input signals. Attaching a thermochromic film is a simple and economical method suitable for converting the existing non-smart windows to the smart windows without any additional control device.

The most promising thermochromic material is Vanadium Oxide (VO_2). It has a sharp phase transition from insulating to metallic phases at 68 °C with a large change in transmittance at near-infrared (NIR) wavelengths before and after the metal–insulator phase transition[2]. A typical high thermochromic VO_2 film tends to be thick and/or complex multilayer structures with brittle property not suitable for a simple past-on film[3]. Embedding VO_2 particles into polymer films can solve this problem and possibly provide better luminous transmittance and thermochromic properties[4].

In this paper, optimization of highly dispersed VO_2 nanowires polymer composite films for maximum thermochromic properties is described using computational simulations. The more comprehensive description of wavelength-dependent thermochromic characteristics of VO_2 nanowire polymer composite films are given in elsewhere[5].

2 Simulation Model

To simulate the light scattering properties of the single VO_2 nanowire, Finite Element Analysis (FEA) modeling by COMSOL Multiphysics was used. In this simulation, scattering cross section C_{scat}, absorption cross section C_{abs}, and angular distribution of scattered light intensity were obtained.

To estimate the optical properties of nanowires composite films using the computed scattering cross section C_{scat} and absorption cross section C_{abs}, geometrical optics calculation was used. To simplify the model, the following assumptions were applied; 1) low fill factor of nanowires with no interactions between the nanowires, 2) aligned nanowires to the composite surface plane that was the worst case for optical transmission, and 3) the perpendicular incident light to the composite surface.

The following Table 1 lists parameters used in computational simulations.

Table 1 Parameter setting for the simulation.

Parameters	Value
Refractive index of the substrate	1.5
Thickness of the composite film	5 μm
Refractive index of the polymer matrix	1.5
Specific weight of VO_2	4.57 g/cm^3
Specific weight of the polymer matrix	1.2 g/cm^3
Nanoparticle fill factor	1 % volume (3.7 Wt %)

This parameter setting was an equivalent of 50 nm VO_2 film[6-7].

Refractive index and extinction coefficient of VO_2 used in this simulation were taken from reference[6]. Index value of 1.5 with zero extinction for a typical polymer was used for the medium. The same index and extinction coefficient as those of the medium were used for the substrate.

3 Results

Fig. 1 shows the scattering and absorption cross sections of various size single nanowires at insulating and metallic phases obtained by FEA simulation.

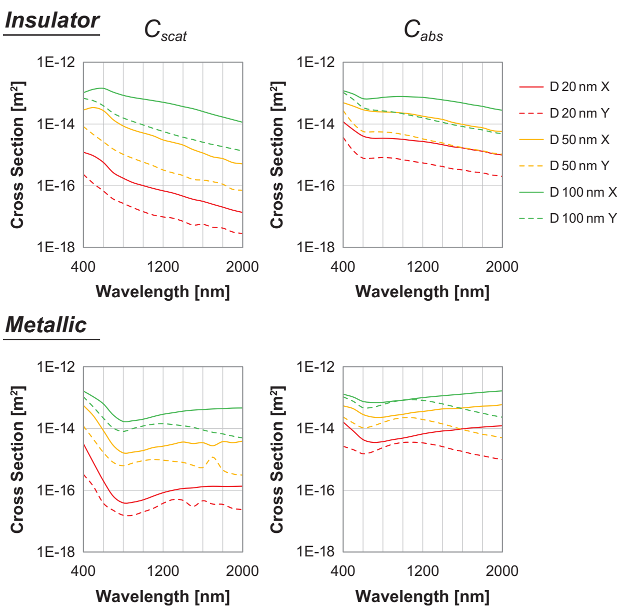

Fig. 1 Scattering and absorption cross sections of various size single nanowires.

The wavelength dependences of the scattering cross section C_{scat} and absorption cross section C_{abs} of Insulating and metallic phases of the single nanowires with 20 to 100 nm diameter and 1000 nm length are shown with X (parallel to long axis of nanowire) and Y (perpendicular to long axis of nanowire) polarization.

Fig. 2 shows the calculated transmittance of 20 to 100 nm VO_2 nanowires polymer composite films for insulating and metallic phases obtained by using the computed scattering cross section C_{scat} and absorption cross section C_{abs} with averaged polarization. VO_2 nanowires lengths were varied from 100 to 1000 nm, and the incident light was assumed to have the same X and Y polarization magnitudes.

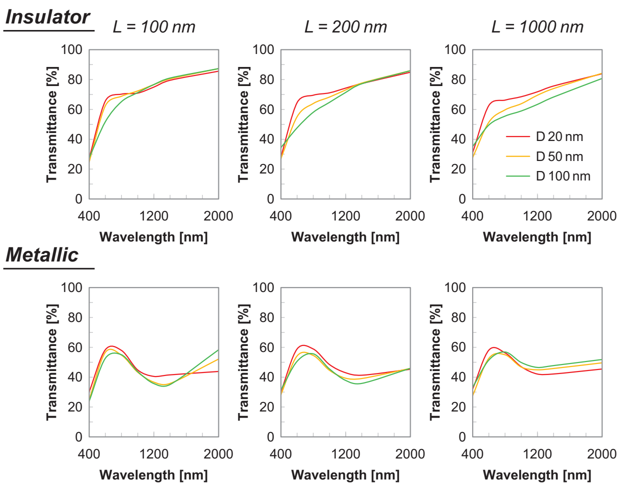

Fig. 2 Transmittance of VO_2 nanowires polymer composite films.

Fig. 3 shows the wavelength dependence of the haze value of various size and lengths VO_2 nanowires polymer composite films for both insulating and metallic phases with the same magnitude of X or Y polarized light.

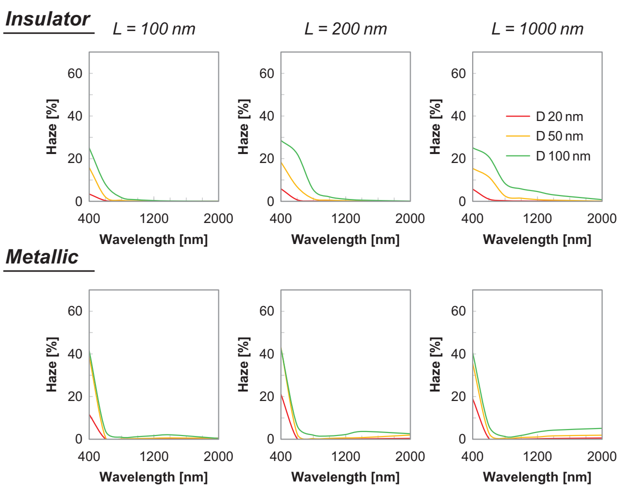

Fig. 3 Haze value of VO_2 nanowires polymer composite films.

4 Discussion

From the computational simulation results, size and length of the nanowires are the key factors to determine the optical properties of the polymer composite films.

Wavelength dependencies of the scattering cross section C_{scat} and absorption cross section C_{abs} are different in insulating and metallic phases. In insulating phase, both C_{scat} and C_{abs} monotonically decrease except for the short wavelength region. In metallic phase, there are small dips for C_{scat} around 800 nm wavelength and for C_{abs} around 600 nm wavelength as expected from characteristics of n and k in metallic phase.

Nanowires have polarization direction dependency as expected due to large differences in X-Y direction aspect ratio. Larger cross sections at X polarized light suggest that long axis of nanowires create strong absorption and scattering effect.

Transmittances of the nanowires composite polymer films in insulating phases exhibit loss of transmittances for larger diameter and longer nanowires films; however, the composite films in metallic phases show large dips in transmittance for larger diameter with shorter length. The polymer composite films with 20 nm nanowires at 100 to 1000 nm length in metallic phase exhibit flat 40 % transmittance above 1200 nm wavelength indicating thermochromic characteristics even for longer IR wavelength.

Using the results shown in Fig. 2, the transmittance difference between insulating and metallic phases $\Delta T = T_{insulator} - T_{metallic}$ (thermochromic efficiency) of various size VO_2 nanowires polymer composite films at 1200 nm wavelength are summarized in Fig. 4. At this wavelength, higher thermochromic efficiencies are achieved by 100 nm long VO_2 nanowires with 20 to 100 nm diameters without considering haze effects.

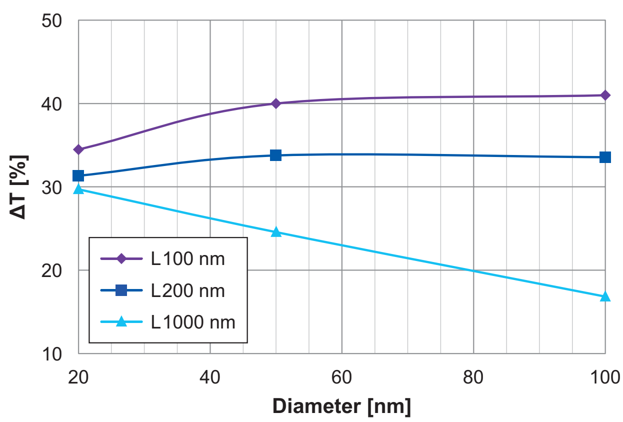

Fig. 4 Transmittance difference between insulating and metallic phases of VO_2 nanowires polymer composite films at 1200 nm wavelength.

In the insulating phase of the nanowires polymer composite films, haze values have no large increase for larger and longer nanowire polymer composite films with hazes starting at about 800 nm wavelength. The metallic phase of the nanowires polymers composite films show no strong wavelength dependency but relatively constant haze values above 600 nm wavelength. It is clear that 20 nm nanowires with 100 nm length polymer composite films provide the best choice from the points of thermochromic efficiency and optical haze.

5 Conclusion

Computer simulation is a powerful tool to investigate optimum structure of VO_2 nanowires in order to obtain high performance thermochromic polymer composite films. Transmittance of the VO_2 nanowires polymer composite films at insulating and metallic phases showed very different wavelength dependence. Nanowires polymer composite films showed a relatively flat transmittance after 1200 nm length. For thermochromic efficiency, 50 nm diameter with 100 nm length nanowires polymer composite films showed the best efficiency. For haze characteristics, the smallest and shortest nanowires polymer composite films exhibited the lowest value. For considering both transmittance efficiency and haze, the best shape VO_2 nanowires were about 50 nm diameter with 100 nm length that produced the polymer composite films with 40 % transmittance efficiency at 1200 nm wavelength and haze vale to about 10 % at 450 nm wavelength.

Acknowledgements

This study was conducted as a part of the collaborative research with Texas A&M University during Yuki Naoi was at Konica Minolta Laboratory USA, The authors acknowledge Prof. Banerjee and his group at Texas A&M University for valuable discussions on thermochromic VO_2 nanoparticles.

References

1) http://www.commercialwindows.org/dynamic.php
2) J. Zhou, Y. Gao, Z. Zhang, H. Luo, C. Cao, Z. Chen, L. Da, and X. Liu, Sci. Rep. **3**, 3029 (2013).
3) P. Jin, G. Xu, M. Tazawa, K. Yoshimura, Applied Physics A **77**, 455 (2003)
4) S.-Y. Li, G. A. Niklasson, and C. G. Granqvist, J. Appl. Phys. 108, 063525 (2010)
5) Y. Naoi, and Jun Amano, J. Appl. Phys. 120, 235301(2016)
6) N. R. Mlyuka, G. A. Niklasson, and C.G. Granqvist, Phys. Status Solidi A 206, 2155 (2009)
7) G. A. Niklasson, S.-Y. Li, and C. G. Granqvist, Journal of Physics: Conference Series 559, 012001 (2014)