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Energimyndighetens titel på projektet – engelska Nanophotonic approaches for efficient colored solar cells	
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Preface

The project ”Nanophotonic approaches for efficient colored solar cells” was funded by the Swedish Energy Agency, Grant No. 49227-1. Midsummer AB was the industrial partner in an advisory role and support in terms of access to their solar cells for testing. KTH Innovation supported the project via creating opportunities to evaluate the project results for potential IP and analysis of commercial interests and needs for colored PV.

Contents

Sammanfattning

Projektet är inriktat på det snabbt växande fältet specialanpassad fotovoltaik, och inom det mer specifikt på färgade solceller för användning i bebyggda områden. I projektet demonstreras innovativa nanofotoniska designer samt tillverkningsmetoder med mål att skapa ytbeläggningar som ger upphov till strukturella färger. Ytbeläggningarna läggs i skiktet mellan sollcell och glas, vilket

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innebär att de kan tillämpas inom en stor bredd av redan existerande solcellsteknik. Titanoxid- och zinkoxidnanodiskar i array-struktur positionerade på eller i glas undersöks genom elektromagnetisk design och simuleringar för att skapa reflekterade strukturella färger i det fotoptiska våglängdsområdet, samtidigt som en hög absorption av solljus bevaras.

De designade array-strukturerna producerar reflekterade strukturella färger i hela det synliga våglängdsområdet med skarpa reflektansstoppar (11-17 nm, FWHM) med hög intensitet (80-100 %). Den reflekterade färgen resulterar i en relativt liten förlust i effektivitet för en Si-solcell: cirka 4-5 % i det blågröna området och cirka 9 % i det röda området för en vertikal infallsvinkel (opolariserat ljus). Polarisationsberoendet visar att för högre infallsvinklar transmitteras det mesta av det p-polariserade ljuset. Således kan förlusten i effektivitet halveras, om man betraktar monteringen av solpaneler i praktiken. Färgernas utseende är mer eller mindre detsamma för betraktningsvinklar upp till ~ 45º och infallsvinklar upp till ~ 70º. De UV-blockerande egenskaperna hos de färggenererande titanoxid- och ZnO-skikten bestäms till > 60 %, vilket är användbart för att minska nedbrytningen av vanliga solcellsinkapslande material.

De tillverkade nanodiskarrayerna (kvadratiska och hexagonala) visar klara färger över hela det synliga spektrumet. Nanodiskarrayerna som tillverkats med toppmodern teknik för deponering, litografi och torretsning skiljer sig från den initiala designen när det gäller form och morfologi. Med anledning av detta visar de uppmätta reflektansspektra mycket breda toppar (inklusive flera toppar) till skillnad från de ideala arrayerna. Reflektansens toppintensitet var hög, 50-80 % för de olika arrayerna. Den uppmätta reflektansdatan stämmer väl överens med detaljerad analys av Mie-spridning från de koniska nanodiskarna inklusive multipolanalys och numeriska simuleringar. De starka färgerna och den höga reflektionen visar tydligt att de tillverkade strukturerna kan användas för andra applikationer inom display och strukturell-färgtryck. Men, ur perspektivet av färgade solceller måste tillverkningsprocesserna utvecklas ytterligare för att erhålla ideala cylindriska nanodiskar med slät ytmorfologi och hög kvalitet i mönstergenerering och överföring. Med förbättrade tillverkningsprocesser kommer färgade ytbeläggningar till solcellsglas att möjliggöra acceptabla förluster i solcellseffektivitet och enorma fördelar till följd av möjligheterna av färgade solpaneler i bebyggda områden.

Summary

The project targets the rapidly emerging field of customized photovoltaics, in particular color solar cells which has a range of applications for the built environment. The project demonstrates innovative nanophotonic designs and fabrication methods to provide structural color coatings on glass/ solar cell interface so that it is applicable for a wide range of solar cell technologies. Titania and ZnO nanodisk arrays on/in glass are investigated by electromagnetic design and simulations for reflective structural colors in the photopic vision regime, keeping the overall sunlight absorption high in the cell. The designed arrays produce reflected structural colors in the full visible range with sharp reflectance peaks (11-

17 nm, FWHM) with high intensities (80-100%). The reflected color from the cover glass results in a relatively small loss in short-circuit current of a Si solar cell: about 4-5% in the blue-green region and about 9% in the red region is determined for vertical incidence (unpolarized light). Polarization dependence shows that for off-angle incidence, most of p-polarized light is transmitted. Thus, the penalty on short circuit current could be smaller by a factor of two, considering practical deployment of solar panels. The color appearance is more or less similar for viewing angles up to ~ 45º and incidence angles up to ~ 70º. The UV-blocking properties of the color generating titania and ZnO layers are determined to be > 60%, useful to reduce degradation of common solar cell encapsulating materials.

The fabricated titania nanodisk arrays (square and hexagonal) display vivid colors across the full visible spectrum. The nanodisk arrays fabricated using the state-of-the-art deposition, patterning and dry etching techniques, deviate appreciably from the design in terms of shape and morphology. Correspondingly, unlike the ideal designed arrays the measured reflectance spectra show very broad peaks (including multiple peaks). The reflectance peak intensity was high, 50-80% for the different arrays. The measured reflectance data is in good agreement with detailed analysis of Mie scattering from the tapered nanodisks including multipole analysis and numerical simulations. The vivid colors and high reflectance peaks clearly show the fabricated structures can be used for other applications in display and structural color printing. However, from the perspective of colored solar cells processes have to be developed further to obtain ideal cylindrical nanodisks with smooth surface morphology and high fidelity in pattern generation and transfer. With improved fabrication colored coatings on solar cover glass will enable acceptable penalties on solar cell efficiency with the huge benefits of deployment of customized colored solar panels in various built environment settings.

Introduction

Photovoltaic (PV) technology has the potential to provide virtually unlimited supply of usable green energy. Economical implementation of PV technology on a global scale requires critical advances not only in materials and devices, but also in functionality to decrease cost and increase power conversion efficiency. The requirement of high efficiency has directed the research on solar cells and several materials, technologies and cell designs are investigated [1,2].

With increased efficiencies, new needs for flexible and so-called color solar cells are emerging, providing new design freedom in architecture. Nanophotonic technology can provide unique properties such as light trapping [3-8]. A very recent development is the emergence of color solar cells mainly driven by potential applications in the built environment. Color solar cells can provide design elements in buildings (window blinds, glass, walls, roof tops (tiles)) from a richer color palette compared to the present black or blue-gray panels. In addition, colored solar cell tiles can be assembled in a modular fashion which offers advantages in artistic design flexibility.

Very few works have addressed color generation in solar cells [9-12]: perovskite solar cells using multi-layer coatings (light interference) for different colors [9]; semitransparent thin film Si solar cells [10]; plasmonic surface layers [11] and partially absorbing Mie scatterers integrated on heterojunction Si solar cells [12]. While most of these approaches have problems with angular dependence of color and/or absorption losses, the dielectric Mie scatterers [12] show the most promising results with narrow band reflected color (green) and only about 10% reduction in short circuit current. However, this was only for green color, the resonator material is absorbs in the visible, and is specific to a particular solar cell (HIT) material/structure. An elegant way is to implement the structural color function in the glass coating or solar cell encapsulant [13]. However, the techniques used for color generation are complex [14,15]. It is an accepted fact that for color solar cells to be viable, it is imperative that the penalty on the cell's efficiency is negligibly small [13,14]. If this requirement is met, the aesthetic function can promote wider deployment of solar cells in the built-environment in a wide range of products.

Outside the solar cell context, dielectric nanostructures are currently being investigated for structural color generation, either in reflection or transmission, for applications in displays and image sensors [16]. The material and geometrical properties of individual nanostructure and their assemblies can provide wavelength selective absorption, transmission or reflection. Efforts to understand light transport, localization and scattering phenomena in assemblies of nanostructures have increased in the recent years [17]. Structural colors in some bird feathers (e.g. cotinga) are due to three-dimensional nanostructures [18]. Such remarkable adaptations provide strong arguments for structural color generation using nanostructures [19]. Recently, sub-wavelength titania resonator arrays on glass substrates produce bright reflected colors by making use of electric (magnetic) dipole interactions with photonic crystal radiative modes [20].

Many of the optical functions discussed above, require direct structuring of the solar cell surface. Considering that much effort is made to surface passivation in solar cell fabrication, a disruptive approach would be to "add-on" functional optical coatings in solar cover glass. Direct embossing of structures using titania and ZnO nanoparticle pastes have been shown [21,22]. However, the procedures require detailed investigations to determine process limits in terms of control over dimensions and spatial separations.

However, providing customized colors in solar cells is challenging in several aspects. Generation of the desired color requires a coating to manipulate the reflected light spectrum which is either integrated on the solar cell or provided in the cover glass/encapsulant. Since solar cells utilize the visible-NIR region of the solar spectrum, there is invariably a penalty on cell efficiency due to loss of photons by surface reflection or by absorption in the coating. For such structural color coatings, the optical design (material layers, nanostructures) should be such that the reflected spectrum is as narrow as possible to minimize the penalty on cell efficiency.

The project address these requirements by considering appropriate materials and fabrication designs. High index metal-oxides like titania and ZnO are transparent in visible region and hence can be used to realize non-absorbing color reflectors. The project focuses on electromagnetic design and simulations of titania and ZnO nanodisk arrays on glass, for narrow and high reflectance in the visible. It demonstrates designs that provide very low reduction in short circuit current in solar cells. In parallel, the project investigates the experimental procedures for fabricating and validating the reflected colors from titania and ZnO nanodisk arrays on glass.

Implementation/Workplan

The project tasks are organized into 3 workparts and are described below.

WP1 Electromagnetic design and modeling

Main objective is to design nanophotonic surface layers for non-iridescent color generation in reflection, keeping the overall sunlight absorption high in the cell. The work focused on Titania and ZnO based nanodisk (cylindrical and tapered shapes) resonator array designs for narrow band (~10 nm) reflected colors in the blue, green and red regions, including polarization independence. Lumerical’s finite difference time domain (FDTD) software is used as the primary tool for design and simulations. In addition, light scattering including multipole analysis was performed for individual nanodisks and in arrays. The work included detailed analysis of reflected color spectra and its impact on solar cell performance and UV blocking properties of titania and ZnO structured coatings. As a figure of merit (FoM) for color solar cell performance, we determine the reflectance of the structural color-layer, weighted with AM1.5. Similarly, FoM of the titania resonator arrays for UV blocking properties was also evaluated.

WP2 Materials and Nanofabrication Methods

Main objectives were to develop fabrication processes for nanodisk arrays on glass (fused silica). We investigated two methods, namely: (a) patterning of thin films (100-200 nm) of crystalline titania, amorphous titania and ZnO and (b) by direct printing of nanoparticles from colloidal suspensions. For process qualifications, test structures were fabricated on silicon for SEM imaging. The thin film fabrication focused on deposition of crystalline and amorphous Titania (TiO_2) and ZnO on fused silica and Si substrates, by different techniques such as magnetron sputtering, sol-gel synthesis and printing from colloidal suspensions. These films were patterned using e-beam lithography and colloidal lithography. Subsequently, nanodisks (arrays) were obtained by pattern transfer by dry etching (thin Cr film was used as the hardmask). The process development included investigations of etching parameters and chemistries in the inductively coupled reactive ion etching system. For direct printing of nanoparticles from colloidal suspensions PDMS molds were prepared using Si-templates. The investigations focused on the limits in terms of pattern fill-factor.

WP3 Optical, material and device characterization

The objectives were to characterize the fabricated titania and ZnO thin films and the nanodisk arrays. This included AFM and SEM measurements for physical dimensions of the structures and roughness. Spectroscopic ellipsometry measurements of the thin films were performed to determine the refractive index in the visible spectrum. The structural colors were determined by reflectance measurements (visible range) using a small-spot set-up for small area patterns defined by ebeam lithography and some large area patterns were separately measured using a spectrophotometer with an integrated sphere. Test c-Si solar cells were measured using a solar simulator.

Results

Resonant Mie scattering from nanodisks engineered for dominant backward scattering result in spectral reflectance peaks, which in the visible generate reflected structural colors. However, Mie resonances typically have low-Q factors and hence the reflected peaks are rather broad (several 10's on nm). In addition, the choice of titania, ZnO or SiNx as materials for the resonators while providing the necessary transparency in the visible have relatively lower refractive index (~1.8-2,5). Thus the index contrast between the resonator and air or glass substrate is low; similarly, for the case when the resonators are embedded in a low-index medium (1.4-1.5). The low index contrast invariably results in weaker scattering and cannot generate vivid colors. Thus, a new physical mechanism is necessary to enhance, spectrally tune and sharpen the reflectance response from such resonators. One of the key phenomena enabling such a functionality relies on the coupling of the Mie resonant modes to the lattice resonance determined by the Raleigh anomaly.

The electromagnetic design and simulation studies demonstrate titania nanodisk arrays can provide reflected structural colors in the full visible range and simultaneous UV blocking. Specifically, by placing the nanodisks in optimized array geometries we show that the spectrally broad scattering profile from Mie scattering is modified dramatically, resulting in narrow band reflectance peaks with reflectance as high as 80-100% for vertically incident unpolarized light. The reflected structural colors in the full visible range are obtained with sharp reflection peaks (11-17 nm, FWHM) and simultaneous UV blocking > 60%. The reflected color from the cover glass results in a relatively small loss in short-circuit current of a Si solar cell: about 4-5% in the blue-green region and about 9% in the red region. These estimates are for vertical incidence and for unpolarized light. In general, the structures optimized for sharp and high reflectance peaks in the red spectral region also show reflectance peaks in the blue.

Detailed investigations on polarization dependence show that the reflected colors are sensitive to the polarization of the incoming light that is incident off-normal or at an angle. For an incident p-polarized light does not produce a reflectance peak for any incidence angles larger than 10º. Whereas, for the case of s-polarized light, the reflectance peak not only exists for all the incidence angles up to 70º, but blue shifts only by about 30 nm. The color appearance, with viewing angles up to ~ 45º, is maintained for incidence angles up to ~ 70º. This also implies that in most practical installation geometries, most of the p-polarized light is transmitted. Thus, the penalty (%) on solar cell short circuit current can be lower by a factor of 2 compared to the estimated values for vertical incidence.

The results presented above are very similar for nanodisk arrays made from (1) sol-gel ZnO (2) crystalline Titania and (3) amorphous titania. The measured average refractive indices are different for these materials and range from 1.9 to 2.5. In each case, it was possible to find the optimized nanodisk size and array parameters. Similarly, both square and hexagonal arrays provide sharp reflectance peaks in the desired spectral regions. However, for the hexagonal lattice the array and disk geometric parameters were found to be less stringent and better optical properties were obtained in the embedded configuration. Especially, the red spectral region is more accessible with suppressed reflectance peaks in the blue-green.

Independently, Mie scattering from nanodisks, individual and in arrays, were analyzed using multipole decomposition to identify the different resonant modes and determine how they contribute to the total scattering. This study was very important to understand the basic mechanisms for color generation and to determine effects of the nanodisk shape. The latter aspect relating to shape arises from the fabrication processes. Fabricated disks invariably deviate from ideal cylindrical disks and additional factors include material's morphology, process induced roughness (surface) and other disorders in pattern definition and transfer. Detailed analysis show that the spectral positions of the resonant modes are sensitive to the sidewall angle of nanodisks. Such shape-specific effects in combination with lattice resonances when placed in arrays result in multiple reflectance peaks. On one hand this suggests possibilities to tune the resonances by shape and may require further optimization of the array geometry to obtain a single well-defined reflectance peak.

The crystalline and amorphous Titania (TiO_2) and ZnO thin films (100-200 nm) were deposited on fused silica by magnetron sputtering. These films were patterned using e-beam lithography. The typical radius varied from 100-150 nm and period from 300-500 nm. Both square and hexagonal arrays were fabricated. The nanodisk arrays were obtained by pattern transfer using dry etching; thin Cr film was used as the hardmask. The process development included investigations of etching parameters and chemistries in the inductively coupled reactive ion etching system. For titania etching, the main chemistries investigated were CHF_3/O_2 and Ar/SF_6; and for ZnO Ar/Cl_2. The optimized etching conditions for each material and chemistry resulted tapered nanodisk profiles. The origin of the tapered profiles is likely due to a dominant physical etching mechanism in addition to possible mask erosion. The narrow spacings between the disks is a major factor causing the lag-effect. Further efforts are necessary to investigate the processing methods to obtain vertical sidewalls.

The fabricated titania nanodisk arrays (square and hexagonal) display vivid colors across the full visible spectrum. However, the measured reflectance spectra invariably show very broad (100-200 nm) peaks, including multiple peaks. The reflectance peak intensity was also high (50-80%) for the different arrays. The measured reflectance data is in good agreement with detailed analysis of Mie scattering from the tapered nanodisks including multipole analysis and numerical simulations. The vivid colors and high reflectance peaks clearly show these structures can be used for other applications in display and structural color printing. However, from the perspective of colored solar cells the broad and high reflectance peaks cause severe penalty on solar cell performance. The main bottle-neck is the technology. Processes have to be developed further to obtain ideal well defined cylindrical nanodisks and high fidelity in pattern generation and transfer. The designed structures do show acceptable penalties on solar cell efficiencies.

In closely related works on Mie resonators and light scattering, we demonstrated enhanced broad-band light absorption in thin film solar cells, polarization dependent color generation from nanostructures,

Discussion

Providing customized colors in solar cells is challenging on how to integrate the color function and how to minimize the penalty due to loss of photons by reflection in the visible region of the solar spectrum. Having the color function in the solar cover glass is a smart solution since it is independent of solar cell manufacturing technologies. There is invariably a penalty on cell efficiency due to loss of photons by surface reflection or by absorption in the coating. Structural colors generated by nanostructured dielectric coatings can provide spectrally controlled reflection while transmitting (scattering) the rest of the light into the solar cell. The penalty on the solar cell efficiency (here, the short circuit current) has to be kept as low as possible, 5-10%, to make colored solar cells attractive for deployment. The project's approach is to use titania or ZnO nanodisk arrays to generate reflected colors - specifically to a produce narrow (10 nm) and high intensity (> 50%) reflectance peak at the desired wavelength. The materials chosen are known for their excellent transparency in the visible and reasonably high refractive indices. In addition, they absorb UV light providing UV-blocking function.
The results obtained from electromagnetic design and simulations clearly demonstrate reflected structural colors in the full visible range from titania and ZnO nanodisk arrays on glass. In particular, sharp reflection peaks (11-17 nm, FWHM) and simultaneous UV blocking > 60% are achieved. A relatively small loss in short-circuit current (5-9%) is determined for vertical incidence (unpolarized light). Polarization dependence shows that for off-angle incidence, most of p-polarized light is transmitted into the solar cell. Thus, the penalty on short circuit current could be smaller by a factor of 2, considering practical deployment of solar panels wherein the incidence angles are invariably off-angle from vertical. The color appearance, with viewing angles up to ~ 45º, maintains for incidence angles up to ~ 70º. The excellent UV blocking property of the coating can prevent degradation of the encapsulation materials, increasing the lifetime expectancy of a solar panel.

Using the state-of-the-art deposition, patterning and dry etching techniques, the fabricated nanodisk arrays deviate appreciably from the design. The fabricated nanodisk arrays show vivid colors, but the reflectance peaks are high but very broad. While these results are attractive for other color applications, for solar cells the high and broad reflectance peaks imply unacceptable penalties on the short circuit current. Our results - experimental and simulations taking into account the actual geometrical and material parameters of the fabricated samples - show that the fabrication of the titania or ZnO nanodisk arrays is indeed very challenging, with stringent demands on shape of nanodisks and on the pattern generation and transfer protocols. Further efforts on the technology is necessary to obtain structures close to the design.

In summary, with improved fabrication colored coatings on solar cover glass will enable acceptable penalties on solar cell efficiency with the huge benefits of deployment of customized colored solar panels in various built environment settings.

Publication list

Publications # 3, 4, 9, 10, 11 deal with the use of Mie resonators (semiconductors, including metal-oxides) and light scattering in the solar cell context, for enhanced broadband light absorption in thin film solar cells and for structural color generation applications. The results are discussed above. In closely related works, we demonstrated polarization dependent color generation from nanostructures (publication #2) and addressed optical anapoles in Mie resonators (publications 6 - 8). The work on optical anapoles provide new insights for light manipulation functions for reflectance, transmittance and light trapping engineering. In publication #1, as a side-aspect bio-inspired structures were investigated for antireflection.

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9. M-E. Kjellberg, A.P. Ravishankar and S. Anand, ”Impact of Resonator Shape and Embedding on the Scattering Characteristics of TiO_2 Nanoarrays”, Optical Materials express – under review (submitted Dec 2024)

Master theses:

10. Richard Berger “Nanophotonic Coatings for Structural Coloration of Solar Cells “, KTH, 2021.
11. Aleksandros Sulaj “Fano Resonances for Reflective Colored Solar Cells “, KTH-SU, 2024

Conference contributions:

12. S. Anand, “Photonic Structures for Light Manipulation Functions in Solar Cells and Light Emitting Diodes” *Invited,* Stockholm Materials hub workshop - Green energy and materials for a sustainable future, Stockholm, 20th October 2022.
13. S. Anand, “Subwavelength Photonic Semiconductor Nanostructures and their Applications”, *Invited,* Photonics Global Conference 2023 (PGC-2023), Stockholm, 21-23 August (2023).
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Bilagor

1. An appendix containing confidential information, unpublished results and proposed continuation plans is included separately. This is not a public report and is not publication on the energimyndigheten’s project database.