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Hybrid photovoltaic-thermal (PVT) solar collectors are being explored to capture the full spectrum of solar energy. Currently, most beam splitting concepts rely on filters that divert photons below the bandgap and/or photons above the bandgap to another solar thermal collector, and the rest for PV conversion. Here, we demonstrate a different strategy: photons of the solar spectrum in the desirable PV band are reflected, while the rest of the photons in the solar spectrum (*i.e.*, below-the-bandgap and high-energy photons) are absorbed in the beam splitter to raise its temperature. The beam splitter also has low emittance in the mid-infrared, effectively suppressing thermal re-radiation losses. The concept is illustrated with a SiO_2/TiO_x multi-layered interference filter deposited on top of a conventional ceramic-metal solar thermal absorber, which has high broadband solar absorptance but low infrared emittance. The resulting quad-band filter with four distinct bands (*i.e.*, high reflectance in the PV band, high absorptance above and below the PV band in the solar spectrum, and high reflectance in the middle- to long-wavelength infrared range) offers a new approach for spectral splitting of photons to harvest the full solar spectrum by combining solar PV and solar thermal systems.

1. Introduction

Currently, two major technologies utilizing the abundant solar energy are the photovoltaic (PV) process that directly converts light into electricity, and the solar thermal process that converts light into heat for hot water systems or for electricity generation via steam turbines[1]. One major limitation of the PV process, however, is that conventional solar cells made from semiconductor materials only convert photons with energy equal to and larger than the material's electron bandgap[2]. Photons with energy below the bandgap are lost. Furthermore, photons with energy greater than that of the bandgap lose the excess energy to heat, leading to efficiency reduction[2]. In order to efficiently utilize the broadband solar spectrum, hybrid photovoltaic-thermal (PVT) technology was proposed to spectrally split the incoming sunlight into several spectral bands, which can be more efficiently utilized by solar cells and solar thermal collectors[3]. Spectrum splitting also reduces the temperature rise in the solar cells and prevents their overheating that further decreases cell efficiency. Our approach to achieve efficient solar spectrum splitting is to develop a quad-band filter that reflects photons with energies just above the PV cell bandgap (defined as the PV band in the solar spectrum) to PV cell(s), while absorbing photons with energies below and above the PV bandgap to be utilized as heat for solar thermal applications. The splitter also has low emittance in the mid- and far-infrared range to suppress its thermal re-radiation losses. Specifically, this paper aims to propose and develop a spectrally selective surface with optical properties shown in Figure 1(a), which directs photons with wavelength from 725 to 1100 nm to Si solar cells, absorbs photons with wavelength shorter than 725 nm and longer than 1100 nm to be utilized as heat in a thermal system, while simultaneously exhibiting high reflectance and thus low emittance in the infrared. For a traditional Si solar cell, the suitable spectral band is approximately 700-1100 nm[4], and 725-1100 nm is chosen as the suitable PV band for our spectral splitter in order to boost the total energy conversion efficiency of hybrid PVT systems[5]. Rest of the solar spectrum is the thermal band. Such quad-band splitters provide potential ways for hybrid PVT system design and offer a compact flat-panel configuration for ease of fabrication and maintenance.
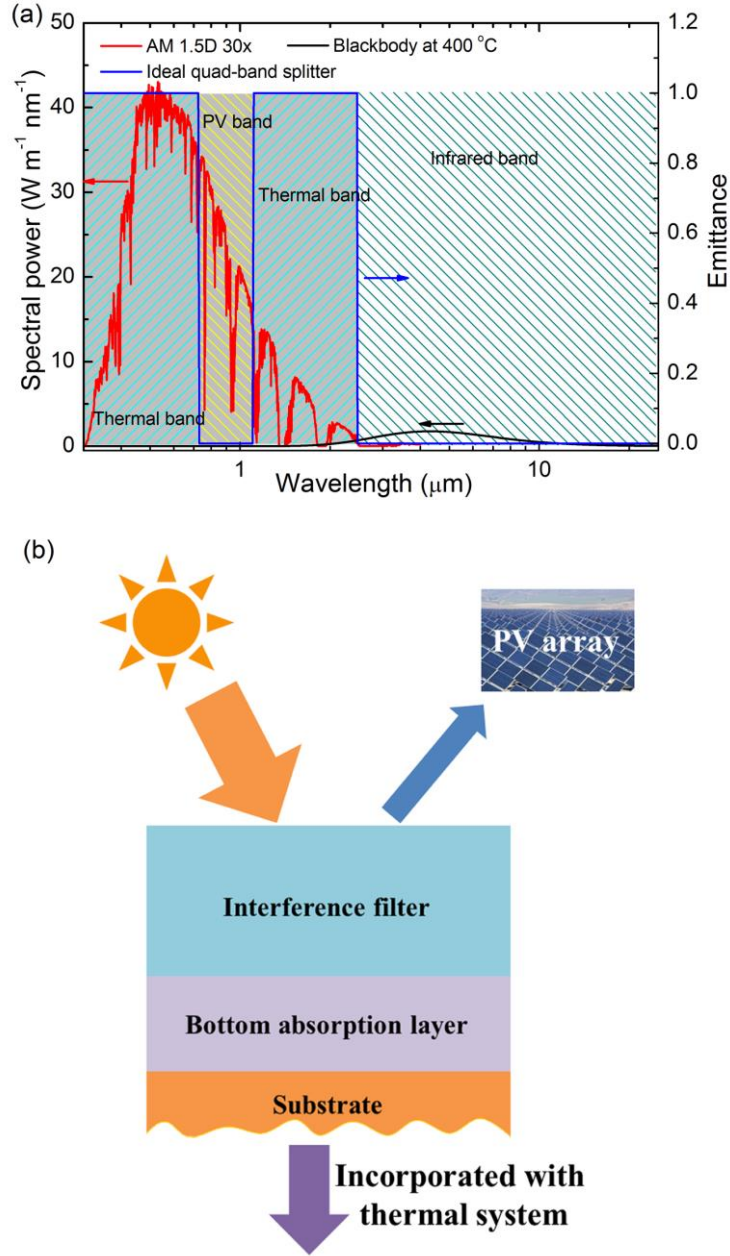
Figure 1. (a) Spectral power of the AM 1.5D solar spectrum at 30x concentration (red) and a spectrum of the 400 °C blackbody (black). The ideal quad-band splitter emittance profile (blue) including two thermal bands (0.3-0.725 μm and 1.1-2.466 μm), a PV band (0.725-1.1 μm for traditional Si solar cells), and an infrared band (2.466-25 μm at the cutoff wavelength of 2.466 μm in this case). (b) Schematic of the proposed PVT system with the solar-spectra quad-band splitter.

Our designed quad-band splitter demonstrates several advantages over existing spectrum splitting methods. For optimum utilization of the entire solar spectrum, existing spectrum splitting strategies often aim to divert photons below or above the bandgap that are not efficient

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for PV operation to a solar thermal system, and photons close to and just-above the PV bandgap to PV cells. A variety of methods, such as optical interference filters[6], selectively-absorbing fluids in combination with PV systems [7], refractive and luminescent surfaces[8], volume and surface holograms[9], dichroic mirrors[8, 10], and luminescent emitters[11] have been utilized for spectral beam splitting. Such systems often require complex optics to direct the short and long wavelength radiation to a thermal component, and additional heat losses resulting from photon absorption in the filter(s) cannot be eliminated. The quad-band splitter combines functionalities of both a splitter and an absorber, which helps to reduce the aforementioned loss from complex optics that integrate a thermal system with the splitter as well as the energy loss from inherent absorption in the splitter.

Dielectric interference filters are suited for the quad-band splitter design due to their sharp transition of optical properties at cut-off wavelengths, flexible design characteristics and low transmission losses[12]. Two types of interference filters commonly used as spectral splitters are distributed Bragg reflectors (DBRs) with alternating layers of high and low refractive index materials and Rugate filters with graded-index coatings.[10] Distributed Bragg reflectors are one-dimensional (1D) photonic crystals (PhCs) that use interference of incident radiation and reflected waves from multiple layer interfaces to form energy band-gaps. They act as mirrors for photons with energies within the bandgaps, enabling frequency-selective filter performance.

In our design as shown schematically in Figure 1(b), quad-band optical characteristics are achieved by integrating triple-band dielectric interference filters widely used as spectral splitters on top of spectrally selective absorbers, which are extensively utilized in solar hot water systems and concentrated solar power plants to capture solar radiation in the form of heat.[13, 14] Cermet based spectral-selective absorbers are integrated in the quad-band splitter owing to their high solar absorptance, low IR emittance as well as good thermal stability for mid- and high-temperature applications.[14] There are many kinds of selective solar-thermal absorbers with different configurations, such as intrinsically selective semiconductor-metal coatings, textured surfaces, multi-layer metal-composites (cermets), and selectively-transmitting coatings on blackbody-like absorbers. Among them, cermet based absorbers such as $Ni-Al_2O_3$, $W-Al_2O_3$, $W-Ni-Al_2O_3$, $Mo-SiO_2$, $W-Ni-YSZ$ [14, 15] can demonstrate a solar absorptance of 0.90~0.96.

The bottom substrate in our design is metal (*i.e.*, mechanically polished copper) so it is compatible with solar thermal systems. The entire structure can be used to direct part of the solar radiation to PV module for electricity generation, and to absorb the remainder of the solar radiation to be used in a solar thermal process. Specifically, we deposit interference filter of SiO_2/TiO_x on top of $W-Ni-SiO_2$ cermet absorber on polished Cu substrate and demonstrate the

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quad-band splitter performance via simulation and experiments. Thermal stability of fabricated structures is also demonstrated experimentally. The resulting quad-band spectral selectivity enables absorption of the full solar spectrum and at the same time reduces thermal re-radiation losses from the absorber surface.

Results and discussion

The optimization of spectral beam splitter is carried out with a simplex gradient search (MATLAB) and the optimized quad-band filters deposited various substrates are summarized in Table 1 (SI, Figure S1).

Table 1. Sputtering parameters of the spectral splitter on selective absorber (WNS), black paint on the mechanically polished Cu substrate, just mechanically polished Cu substrate, or glass slide.

Sample	Bottom absorption layer	Spectral beam splitter (thickness of each layer (nm))								
		SiO_2	TiO_x	SiO_2	TiO_x	SiO_2	TiO_x	SiO_2	TiO_x	SiO_2
SST-1	Varies*	67	102	169	100	171	100	170	80	72

SiO_2 : sputtered with a RF power density of 4.4 W/cm^2.

TiO_x: reactive sputtering of Ti with a DC power density of 6.6 W/cm^2 and a mixture plasma environment of Ar and O_2 (oxygen partial pressure: 0.6 mTorr, argon partial pressure: 2.4 mTorr).

Varies*: WNS, paint on mechanically polished Cu substrate, just mechanically polished Cu substrate, and glass slide.

Figures 2(a) and 2(b) show the optical properties of samples with SiO_2/TiO_x interference filters deposited on different substrates and absorbers in the solar spectrum (200-2500 nm) and infrared spectrum (2.5-25 μm), respectively. Various absorbers on different substrates were used. We show in the following sections that W-Ni-SiO_2 (WNS) cermet selective absorber on copper substrate is the best substrate for the designed quad-band splitter. As indicated in Figure 2(a), strong reflectance in PV band (725-1100 nm) from the top interference filter primarily tailors shape of the quad-band splitter's spectral selective response in the solar spectrum. Characteristics of band splitting can be observed clearly for samples on WNS-Cu, Paint-Cu and

glass substrates with high reflectance in the wavelength range of 725-1100 nm which can be directed to a PV module for direct electricity generation, and low reflectance in the rest of the solar spectrum for thermal process. In the infrared range, strong infrared reflectance and low emittance is observed for the splitter deposited on WNS-Cu and Cu substrates, with narrowband absorption at ~9.6 μm (Figure 2b), which originates from the Si-O-Si stretching vibration in SiO_2.[16] It is thus evident that the stack with SiO_2/TiO_x filter deposited on W-Ni-SiO$_2$ cermet selective absorber demonstrates the desired properties of the quad-band spectral splitter.
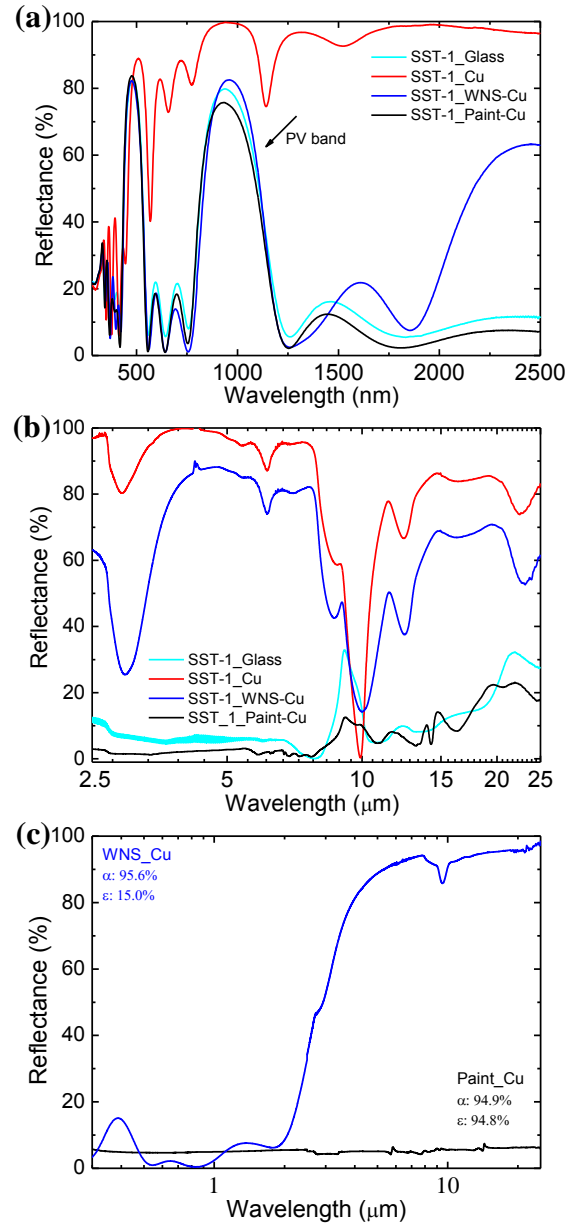

Figure 2. Reflectance characteristics of SST-1 quad-band splitter using SiO_2/TiO_x as an interference filter deposited on glass (cyan, SST-1_Glass), copper (red, SST-1_Cu), WNS absorber on Cu (blue, SST-1_WNS-Cu), and paint on Cu (black, SST-1_Paint-Cu) substrates in

the wavelength range of 0.2 to 2.5 μm (a), and in the wavelength range of 2.5 to 25 μm (b). All the quad-band splitters have the same thickness for the SiO_2 and TiO_x layers. (c) The reflectance spectra of a W-Ni-SiO_2 cermet (WNS) and a black paint on the mechanically polished Cu substrate.

Dielectric filters deposited on glass and Cu substrates are shown in comparison in Figure 2(a) and 2(b) as well. The sample deposited without absorber layer on copper substrate (SST-1_Cu) demonstrates high reflectance across the entire solar and infrared spectra, except for low reflectance in UV and around 9.6 μm, which correspond to strong absorption of Cu and SiO_2 respectively. Since all photons transmitted through the filter are reflected by the Cu substrate, and not absorbed, the sample on Cu substrate consistently shows high reflectance. The stack on the glass slide (SST-1_Glass), which has a similar configuration with a conventional interference filter, shows low reflectance in the thermal band indicating high transmittance, not absorptance. Photons transmit through the SiO_2/TiO_x stack and the glass substrate without being absorbed due to the lack of absorber layer, therefore additional optics is needed to direct the transmitted photons to a thermal absorber. Figure 2(b) also shows low reflectance for samples deposited on glass due to the strong absorption and high emittance of glass in the infrared.

The stack on the paint-coated Cu substrate (SST-1_Paint-Cu) also has good reflectance in the PV band. However, absorptance in the thermal band is nonselective with high absorptance both in solar spectra range excluding the PV band, and in the IR band (Figure 2b), leading to high emittance. Such difference in infrared properties can be attributed to different properties of the bottom absorbers. Figure 2(c) compares reflectance characteristics of the two different absorbers, W-Ni-SiO_2 cermet absorber and commercial black paint, both coated on mechanically polished Cu substrates. Both absorbers demonstrate low reflectance below 2 μm as shown in Figure 2(c), which indicates high solar absorptance, while WNS absorbers demonstrate selective solar absorption with lower emittance in the infrared and commercial black paint exhibits non-selective absorption with high absorption and thus emittance in infrared region. The solar absorptance (α) and total-directional emittance (ε) can be estimated by the weighted integration of the reflectance spectra with solar spectrum (AM 1.5 direct + circumsolar), and weighted integration of the reflectance spectra with the blackbody spectrum at 400 °C, respectively. The WNS absorber displays spectrally-selective absorption with solar absorptance of 95.6% and total-directional emittance of 15.0%. However, the paint shows non-selective absorption with

almost the same solar absorptance and total-directional emittance. Both can act as the absorption layer due to their superior solar absorptance (~95%), nonetheless the high emittance of the paint indicates high thermal loss when operating at high temperatures, especially under moderate optical concentrations. We thus conclude that the WNS absorber should be used for the proposed spectrally-selective absorber-splitter. With the benefit of W-Ni-SiO$_2$ cermet spectrally selective absorber (WNS), quad-band splitting can be achieved in the structure deposited on the WNS absorber-coated Cu substrate (SST-1_WNS-Cu) with low absorptance in the PV band and the IR band, and selective high absorptance in the thermal band. The increased reflectance in the wavelength range beyond 2 μm compared to the stack on paint-coated copper substrate indicates low thermal re-emission loss and potential applications in PVT hybrid solar collectors.

The designed reflection window can be shifted by tuning the thicknesses of the individual layers in the top interference filter. We achieve a reflection window in the visible range with SiO$_2$/TiO$_x$ stack by decreasing the thickness of each layer (SI, Table S1 and Figure S2) and the remaining part except the thermal IR range still keeps low reflectance, indicating high photothermal conversion efficiency due to high solar absorptance and low emittance.

Since the proposed quad-band splitter will be also used as the thermal absorber, thermal stability at operational temperature is crucially important. Thermal annealing is carried out on those samples in vacuum at 400 ^{o}C for 3 days. The solar spectrum splitter on Cu substrate (SST-1_Cu) has copper color and other two on WNS absorber and paint-coated Cu substrates are blue (SI Figure S3). The appearances of those samples on Cu and WNS absorber-coated Cu do not change too much upon annealing. However the coatings on the paint-coated Cu substrate (SST-1_Paint-Cu) partially peel off from the substrate after annealing, likely due to the mismatch of the coefficient of thermal expansion among the Cu substrate, the paint absorption layer, and the top multilayer TiO$_x$/SiO$_2$ stack. Thus we only investigated the reflectance response and morphology change of SST-1_WNS-Cu upon annealing due to the lack of the splitter spectral characteristics in SST-1_Cu and the bonding issue in SST-1_Paint-Cu after annealing. As shown in Figure 3(a), SST-1_WNS-Cu displays almost identical reflectance characteristics between the pristine one and the annealed one indicating the solar spectrum splitter is pretty stable and can survive up to 400 ^{o}C in vacuum, due to the good stability of W-Ni-SiO$_2$ cermet layer in the WNS absorber. This can be further confirmed by the smaller surface morphology change upon annealing (Figure 3c) in comparison with that of the sample before annealing (Figure 3b). The root mean square roughness (R_q) of the sample changes from 10.8 nm to 10.6 nm upon

annealing, and particle sizes in the sample remain almost unchanged before and after the thermal annealing.
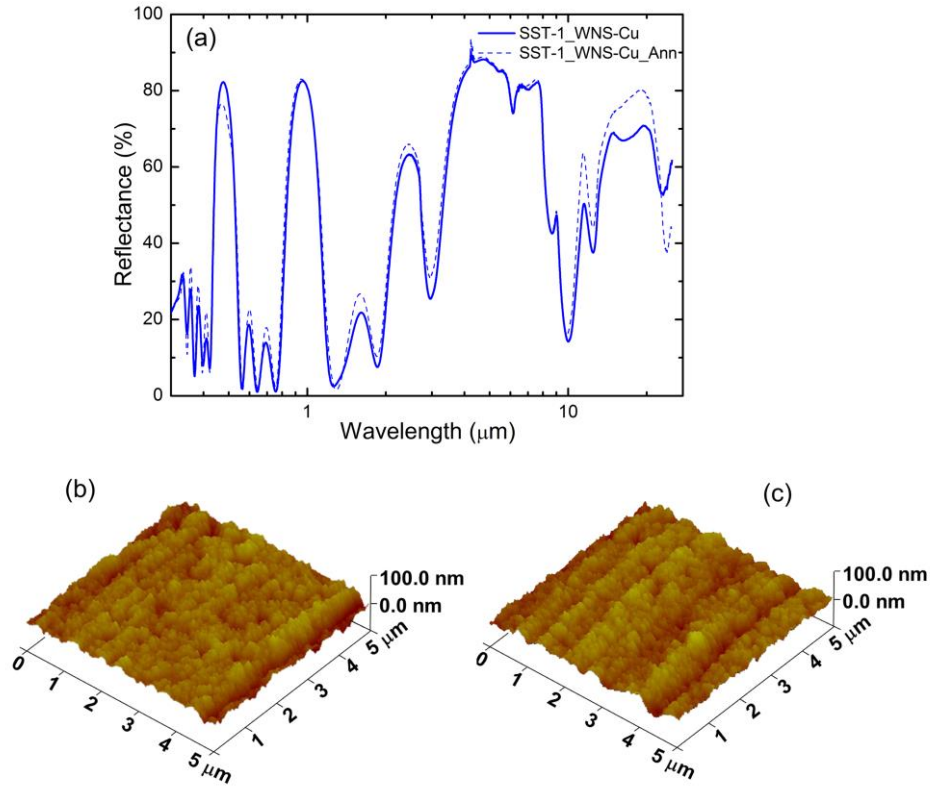

Figure 3. (a) Reflectance characteristics of the spectral splitter (WNS coated-Cu substrate, SST-1) before (solid line) and after (dash line) annealing at 400 ^{o}C for 3 days in vacuum. AFM images of SST-1 before (b) and after (c) the annealing of 3 days at 400 ^{o}C.

Although we have experimentally demonstrated the concept of the spectral absorber-splitter with selective solar absorptance as well as selective absorptance in the thermal IR band, the reflectance in the PV band is not high enough and the highest reflectance for the SST-1_WNS-Cu sample is ~82% at the wavelength of 956 nm resulting from the few number of interference layers in the multilayer SiO_2/TiO_x stack compared to other reported interference filters.[6] Further tuning of the layer thickness and number of layers, and the dielectric constant will be the focus of future studies. In another hand, the low solar weighted reflectance in PV band means small fraction of solar radiation can be directed to PV module and huge contribution on thermal band. What the PV band and thermal band efficiencies should be depends on the real requirements. If we want more radiation to PV band, the solar weighted reflectance in PV band should be increased, rather if more radiation to thermal band, the solar weighted reflectance in thermal band should be decreased.

Conclusion

In conclusion, we proposed a quad-band solar spectral splitter with both spectral splitting and solar absorption functions, which also suppresses radiative heat loss. The splitter is integrated on a single planar substrate, and is based on a combination of a multilayer interference filter and a selective solar thermal absorber. We experimentally demonstrated the quad-band splitting characteristics with SiO_2/TiO_x multilayer stacks deposited on the W-Ni-SiO_2 (WNS) selective absorption layer on Cu substrate. The quad-band solar spectrum splitter with WNS absorber exhibits good stability up to 400 ^{o}C in vacuum. The proposed quad-band solar spectrum splitter also has the potential to be integrated with devices utilizing visible light, such as a visible light-driven plasmonic photocatalyst.[17] The overall conversion efficiency of the system would be boosted resulting from the full utilization of the remaining part of solar radiation spectrum for a thermal process.

Experimental Section

Detailed experimental part is in the supporting information.

Supporting Information

Supporting Information is available online from the Wiley Online Library or from the author.

Acknowledgments

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