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Research Paper

## Hollow photonic structures of transparent conducting oxide with selective and tunable absorptance

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## ABSTRACT

We adopted hollow 3D structures of transparent conducting oxides (TCO) for efficient emission control. TCO has high transmittance in the solar spectrum and tunable optical properties in the infrared regime due to their plasmonic property. The IR emissivity can be further adjusted by the geometry. Here, using solid and hollow triangles as an example, we modeled the spectral absorptance of 3D TCO structures with various carrier concentrations and geometrical factors. We showed that hollow triangular structures enhance the spectrally selective absorption, namely, high emittance in IR and low absorptance in the solar spectrum. This is because the large primary sizes of the triangles can interact strongly with the longer wavelength mid-IR while the small wall thickness of the hollow structures reduces the overall absorption volume for the shorter wavelength light in the near-IR and visible regimes. Further, by changing their angles, the hollow features can be used to tune the IR emissivity within a large range (from 0.14 to 0.8). The selective and tunable absorptance of 3D hollow structures of TCO may find applications in passive radiative cooling, solar thermal absorbing, and tunable windows glazing.

## 1. Introduction

Controlling thermal emission in the mid infrared (MIR) range is of great interests for thermal engineering with broad applications ranging from radiative cooling of spacecrafts [1] and terrestrial objects [2,3] to building envelopes [4], and from solar-thermal absorber [5,6] to thermal camouflage [7] and heat shielding (e.g., white roof [8]). In many of these applications, it is often desirable to have selective absorptance in different parts of the spectrum. For example, for many solar utilization devices (such as solar-thermal, thermoelectric, and photovoltaic) [6,9–12], absorption in the solar spectrum is desirable for harvesting solar energy to heat or directly into electricity. On the other hand, for cooling application, it is preferred to have low solar absorption and high infrared emission [2,3,13,14]. Achieving high spectral selectivity is a key strategy to enhance the figure of merit of the thermal absorbers or emitters [15,16]. Therefore, these two opposite applications have adopted different materials, e.g. metals for solar-thermal absorbers and glass for radiative emitters [2,3,11,17]. Plasmonic materials based on surface phonon polaritons (SPhP), such as SiO<sub>2</sub> and SiC, have been traditional materials of choice to engineer the thermal emission, because its resonant frequency is located at thermal wavelength (~10 μm). SPhP also has much lower optical loss in the MIR

compared to surface plasmon polaritons (SPP), which is attractive for some applications such as IR detection [18], where high quality factor is needed. However, for certain applications in thermal absorption/emission, such as solar absorbers and radiative cooling emitters, high optical loss and high extinction coefficient can be taken as an advantage. From this perspective, plasmonic metal structures are particularly suitable. In addition to spectral selectivity, the ability to dynamically tune thermal emission is also desirable in many thermal engineering applications, such as thermochromic windows [19,20] and thermal camouflage. Many of these features are realized by metal-insulator phase transition of VO<sub>2</sub> [4,21,22] and related oxide materials [23], as well as conductive polymers [24].

Recently, aluminum-doped ZnO (AZO) and other transparent conductive oxides (TCO) have emerged as an interesting plasmonic material with both spectral selectivity and dynamic tunability. Due to its plasmon frequency residing in the near-IR (NIR) regime, TCO is dielectric (transmitting) in the solar/visible spectrum and metallic (both reflective and absorbing) in the IR spectrum. The cutoff wavelength is determined by the plasmonic frequency described by the Drude model [25–27]. This feature offers a large flexibility when designing thermal infrared structures. Most notably, when the highly reflective feature of the metallic state is used, TCO can be used as a high-transmittance and

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low-emissivity coating [28]. This coating can be directly applied onto glass to make low-emissivity windows [29] or deposited on a black solar absorber to make spectrally selective solar collectors [30]. AZO has been used for various applications and hence has been prepared with a wide range doping concentrations from  $\sim 10^{19} \text{ cm}^{-3}$  to  $10^{21} \text{ cm}^{-3}$  by various techniques [25,31–34]. In particular, atomic layer deposition (ALD) has been used to achieve high carrier concentration up to  $\sim 1 \times 10^{21} \text{ cm}^{-3}$  [25]. Also, Ga co-doping can be used to additionally increase the doping level beyond the solubility limit of Al in ZnO [31]. Therefore, the doping concentrations studied here (up to  $\sim 5 \times 10^{20} \text{ cm}^{-3}$ ) are well within the experimentally accessible regime.

While TCO was originally used for NIR due to its low loss in this spectrum compared to pure metals [35,36], it is also becoming interesting for MIR applications precisely due to its high loss in this spectral regime, i.e., the absorbing feature of metallic TCO. The infrared properties again can be controlled by tuning the plasma frequency, which can be designed either during the material preparation or dynamically tuned electrostatically. The IR absorption or emission properties can be further engineered by the structures. Various types of photonic structures [37,38] have been developed in the forms of periodic nano/microstructures and multilayers made of metals and/or dielectrics. For instance, it was recently discovered that three-dimensional (3D) photonic structures of hairs of Saharan silver ants are effective for passive radiative cooling [39,40] with high emittance in the MIR but high reflectance in the visible and NIR regimes. In addition, metamaterials and metasurfaces of TCO have also been utilized to control the IR properties. For instance, it was shown recently that thin metasurfaces of AZO can possess both high IR emissivity ( $\sim 0.8$ ) and low absorptance in the solar spectrum ( $\sim 0.16$ ) [1], which is useful for radiative cooling of spacecraft. Owing to the extensive experimental studies of 3D structure with various applications in photonic crystals, drug delivery, soft robotics and so on., the complex structures can be fabricated using dissolvable template [41–43] or 3D printing techniques [44,45].

In this study, we adopted 3D photonic structures of AZO and compare the emissive behavior at room temperature with common bulk blackbody,  $\text{SiO}_2$ . We performed optical modeling to show the effectiveness of tuning the visible and MIR spectral properties independently using both the carrier concentration and structural changes. In addition, with the benefits of the hollow 3D structures, we demonstrated tunable MIR emittance and low solar absorptance.

## 2. Calculations

We started from the permittivity ( $\epsilon$ ) of AZO. It is well known that the dominant free carriers make the  $\epsilon$  of AZO follow the Drude model over the broad energy range, except the high energy in UV region ( $\sim 300 \text{ nm}$  in wavelength) due to inter-band absorption (ZnO bandgap energy is 3.37 eV, or 369 nm), and the relation can be described as [1]:

$$\epsilon(\omega) = \epsilon_b - \frac{\omega_p^2}{\omega^2 + i\omega\Gamma} + \frac{f_1 \omega_1^2}{\omega_1^2 - \omega^2 + i2\omega\Gamma_1} \quad (1)$$

$$\omega_p^2 = \frac{ne^2}{m^* \epsilon_0} \quad (2)$$

where  $\epsilon_b$  is permittivity at infinite frequency,  $\epsilon_0$  is permittivity of vacuum,  $\omega_p$  is plasmon resonance frequency,  $\Gamma$  is damping coefficient followed by the Drude model,  $f_1$  is amplitude,  $n$  is carrier concentration, and  $m^*$  is effective mass of an electron.  $\omega_1$  is spectral position and  $\Gamma_1$  is damping constant at optical bandgap which follows Tau-Lorentz relation. We can set the resonance frequency out of the dominant solar spectrum to make AZO optically transparent within the solar or visible spectrum.

We then used finite element method (FEM) to simulate the absorptance of 3D triangular structures of AZO and  $\text{SiO}_2$ , as shown in Fig. 1(a). Here the triangular structure is used as an example to show

the effectiveness of tuning the photothermal properties of 3D AZO structures, and similar ideas can be applied to other structures. We varied the geometry of the triangular structures as well as the carrier concentration in AZO. Perfect conductor was used as a back reflector so as to evaluate the emission and absorption purely from the material of interest (i.e., AZO).

To achieve spectral selectivity, e.g. low absorptance in the solar/visible spectrum and high emittance in the blackbody spectrum at 300 K, we used two different length scales: skin-depth ( $\delta$ ) and wavelength. For high absorptance in MIR region, structure sizes comparable to wavelengths are required to scatter the infrared, leading to longer effective optical path for stronger light absorption. On the other hand, it is necessary to have the sizes smaller than the  $\delta$  of shorter wavelength lights to enhance the transmittance in the visible regime. These two requirements are often contradicting to each other. Even though AZO is dielectric, i.e., non-absorbing, for photons with wavelength shorter than the plasmon wavelength, the structures used for enhancing the IR absorption would still introduce the scattering effect in the visible regime. To mitigate this issue, we used 3D hollow triangular structures, as shown in Fig. 1(a). The hollow structures have the primary length scale described by its period ( $P$ ) comparable to the wavelength of infrared (i.e., a few microns), so as to enhance the scattering and absorption in the MIR regime. In the meantime, the hollow structures have very thin wall, with its thickness ( $h$ ) much smaller than the skin depth  $\delta$  of shorter wavelength light for high visible transmittance. This design can decouple the opposite requirements on the length scales for visible transmittance and IR absorptance.

To illustrate this idea quantitatively, we computed the distribution of electric field intensity in a representative hollow triangular structure, as shown in Fig. 1(b). A plane wave was used to model the normal incident light, and the figure shows an example of a triangular structure with  $P$  of  $10 \mu\text{m}$  and  $h$  of  $300 \text{ nm}$ . As expected, the thickness  $h$  is much smaller than the skin depth  $\delta$  ( $\sim 10 \mu\text{m}$ ) of AZO in the Vis regime, so there is little absorption of the incident light but only reflection and refraction at the air/AZO interface due to refractive index mismatch. On the other hand, there is enhanced light scattering and trapping in the MIR, thereby we observe the high field intensity confined between neighbor triangular structures. Also, the distinct intensity difference between the outer and inner spaces of the triangle implies significant light absorption inside the hollow structure. This is due to the multiple reflections by the 3D structure and the back reflector. Here we conducted modeling with various cases to achieve spectral selectivity as shown in Fig. 1(c), and the results is discussed in Section 3. We aimed to achieve a cutoff wavelength near  $3 \mu\text{m}$  that separates the solar (AM0) and blackbody spectrum at 300 K [Fig. 1(c)].

## 3. Results and discussion

As shown in Eq. (1), permittivity  $\epsilon$  of TCO is controlled by the carrier concentration. Fig. 2(a) and (b) displays the real ( $\epsilon'$ ) and imaginary ( $\epsilon''$ ) part of  $\epsilon$  ( $\epsilon = \epsilon' + i\epsilon''$ ), respectively. The plasmon frequency is determined when  $\epsilon' = 0$ . As expected, as the carrier concentration increases from  $5 \times 10^{19} \text{ cm}^{-3}$  to  $5 \times 10^{20} \text{ cm}^{-3}$ , the plasmon wavelength is shifted from  $\sim 8 \mu\text{m}$  to  $\sim 2 \mu\text{m}$ . In addition, higher carrier concentration also results in a larger  $\epsilon''$  at the same wavelength, leading to higher absorption and shorter skin depth  $\delta$ , as shown in Fig. 2(c). We also compared the  $\delta$  to those of  $\text{SiO}_2$  in MIR (Fig. 2(c)). As mentioned earlier,  $\text{SiO}_2$  is widely used as an MIR emitting material due to the SPhP resonance within the Reststrahlen band (8–9.5  $\mu\text{m}$ ). As shown in Fig. 2(c), within the narrow Reststrahlen band,  $\delta$  of  $\text{SiO}_2$  is comparable to that of AZO. However, outside of this range,  $\text{SiO}_2$  possesses much larger  $\delta$  over the broad range. This result indicates that AZO can be a more effective MIR emitting material, especially for a wide range of emitter temperature.

Besides the carrier concentrations, we can also tune the photothermal properties with the structures. In order to show the separate

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