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Abstract. In a thermophotovoltaic (TPV) system, a heat source brings an emitter to incandescence and the spectrally confined thermal radiation is converted to electricity by a low-bandgap photovoltaic (PV) cell. Efficiency is dominated by the emitter's ratio of in-band emissivity (convertible by the PV cell) to out-of-band emissivity (inconvertible). Two-dimensional photonic crystals (PhCs) offer high in-band emissivity and low out-of-band emissivity at normal incidence, but have reduced in-band emissivity off-normal. According to Lambert's law, most thermal radiation occurs off-normal. An omnidirectional PhC capable of high in-band emissivity at all angles would increase total in-band power by 55% at 1200°C. In this work, we present the first experimental demonstration an omnidirectional hafnia-filled 2D tantalum PhC emitter suitable for TPV applications such as combustion, radioisotope, and solar TPV. Dielectric filling improved the hemispherical performance without sacrificing stability or ease of fabrication. The numerical simulations, fabrication processes, and optical and thermal characterizations of the PhC are presented in this paper.

The increasing power demand of communication equipment, sensors, micro-robotic platforms, and portable electronics has driven recent interest in micro- and millimeter-scale thermophotovoltaic (TPV) generators. Thermophotovoltaic energy conversion is a solid-state high-temperature heat-to-electricity conversion scheme with no moving parts that allows for scalable energy production with high specific energy from a variety of heat sources. In TPV systems, thermal radiation from a heat source at high temperature drives a suitable low-bandgap photovoltaic cell (PV). The heat can be produced by hydrocarbon combustion, ideal for lightweight, high specific energy portable power sources [1]; by radioisotope decay, ideal for space missions and remote missions requiring power sources with long lifetimes and low maintenance [2]; or by concentrated solar radiation [3]. A selective emitter between the source and the cell enables high heat-to-electricity conversion efficiency by enhancing in-band (convertible) radiation and suppressing out-of-band (nonconvertible) radiation that would otherwise be wasted. TPV has only recently been enabled by advances in two areas: low-bandgap III-V semiconductors [4] and high-temperature photonic crystal selective emitters [5]. A viable TPV selective emitter requires: high temperature stability for a long operational lifetime, good optical performance, and a simple fabrication process capable of producing large area samples. Most of the selective emitters currently available (fabricated as 1D, 2D, and 3D photonic crystals, metamaterials, as well as from natural materials) only meet one or two of

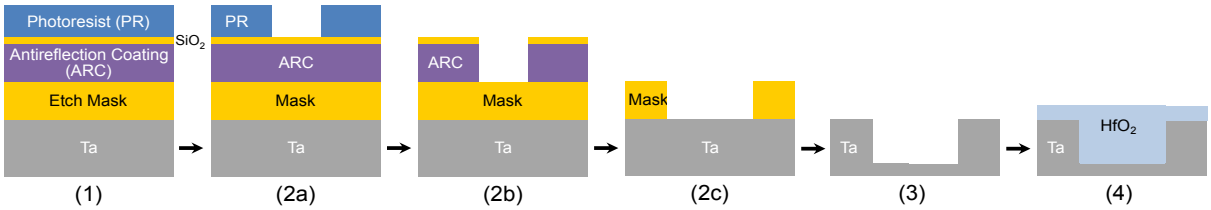

Figure 1: A general process flow for the fabrication of our 2D PhC involves (1) pattern generation, (2) pattern transfer into a hard mask by etching, (3) pattern transfer into the metallic substrate by etching, and (4) filling of the cavities with HfO_2 by ALD.

these criteria. Indeed, while multilayer stacks [6], cermets [7] emitters are easy to fabricate, heterogeneous platforms are subject to thermo-mechanical stresses and chemical reactions at material interfaces that are initiated at elevated temperatures. Even homogeneous material platforms, while not being subjected to stresses, can degrade at high temperature: radius of curvature driven surface diffusion shortens the lifetime of complex structures such as 3D photonic crystals [8]. Our 2D metallic photonic crystal meets all of the requirements of a selective emitter for high-temperature energy conversion.

Our phonic crystal consists of a square periodic array of holes etched into the low-emissivity polished metallic substrate, polycrystalline tantalum. A refractory metal is used as the substrate for the photonic crystal due to its high melting point, low vapor pressure, advantageous low emissivity in the infrared, and ability to be etched. The emissivity of the substrate material is selectively enhanced to near that of a blackbody emitter by the resonant cavity modes resulting from the etched holes. The spectral range of enhancement can therefore be easily tailored to the specific system needs by tuning cavity geometry to match the photonic crystal to different low-bandgap PV cells. A conformal passivation layer of a dielectric material, hafnium dioxide (HfO_2) is used [9] to further achieve high-temperature stability and prevent structural and optical degradation after long operational lifetimes.

The performance of our conformally coated photonic crystal has been consistently good at normal incidence [9, 10], however most of the thermal radiation actually occurs off-normal according to Lambert's law. While metamaterial designs based on metal-dielectric stacks [11] and 2D metallic pyramid arrays [12] have shown promise, they are difficult to fabricate and have not been experimentally demonstrated at high temperatures under extended operation. A simple approach of filling a 2D metallic photonic crystal with a dielectric was introduced [13] to obtain omnidirectional, polarization insensitive, wavelength selective thermal emission. In this paper, we present the first *experimental* demonstration of a HfO_2-filled 2D tantalum photonic crystal selective emitter that further increases TPV system efficiencies by increasing total in-band power (at all polar angles and polarizations) without sacrificing ease of fabrication.

In our 2D photonic crystal selective emitter, the high emittance at short wavelengths is achieved by matching the radiative rate to the absorptive rate for each resonance in the square periodic array of holes. This is assisted by the fact that both the radiative and absorptive rates increase with mode number and hence frequency. At long wavelengths the emittance is due to a surface area weighted effective impedance created by the holes [14]. The result is near-blackbody emittance at short wavelengths as well as emittance almost as low as that of polished tantalum at long wavelengths, with a sharp cutoff between the two regimes. For off-normal incidence however, this performance significantly drops [13]. The spectral emissivity of the photonic at normal incidence and the hemispherically-averaged emissivity for the photonic crystal structure was simulated by the Fourier Modal Method using a freely available software package [15]. The material properties of the substrates were taken into account using a Lorentz-Drude model fitted to the elevated temperature emissivity to capture the optical dispersion of the substrate at high temperature. Nonlinear optimization determined the period, cavity radius, and height with bounds based on the desired cutoff wavelength and other fabrication constraints.

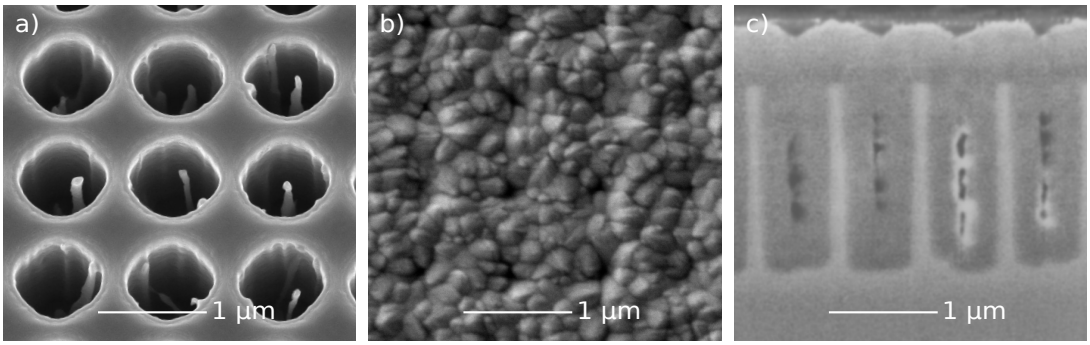

Figure 2: SEM micrographs of the photonic crystal (a) before and (b) after filling of the cavities with HfO_2 by ALD. (c) Cross section after filling.

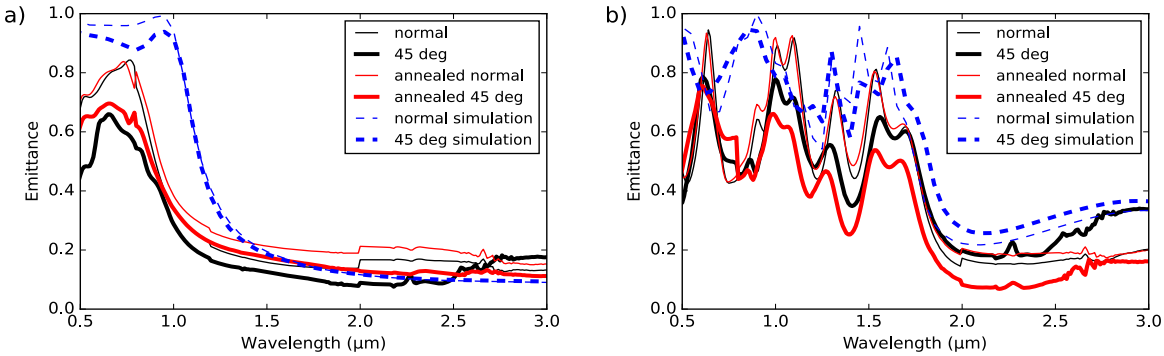

Figure 3: Normal and 45° incidence measurements (before and after anneal at 1000°C for 24 hours) of the (a) unfilled and (b) HfO_2-filled photonic crystals compared with their normal incidence and hemispherically averaged simulations.

The photonic crystal was fabricated in the same way as in our previous work [16]. The fabrication process began with by defining a periodic pattern in photoresist (PR) by interference lithography. Two orthogonal exposures in a Mach-Zehnder setup generated the pattern. A numerically optimized anti-reflection coating (ARC)/SiO_2/PR stack was used where the SiO_2 served as a protection layer for the ARC while etching the PR. Using reactive ion etching (RIE), the pattern was transferred through the tri-layer stack into the SiO_2 mask deposited by plasma-enhanced chemical vapor deposition (PECVD). The tantalum was etched by deep reactive ion etching (DRIE) using a Bosch process. After the final pattern transfer, the residual passivation layer was removed by oxygen plasma and the residual SiO_2 mask was removed by hydrofluoric acid. The fabrication steps are described in Figure 1. The final step - the filling of the photonic crystal cavities by atomic layer deposition (ALD) - was done in the same way as when depositing the conformal layer, but simply depositing for a longer period of time until the cavities are filled.

The fabricated photonic crystal was measured by scanning electron microscopy (SEM) and found to have a period $a \sim 0.5$ µm, radius $r \sim 0.2$ µm, and cavity depth $d \sim 1.5$ µm before filling, as shown in Fig. 2 (a). The surface of the photonic crystal after filling is shown in Fig. 2 (b). The filling of the holes was found to be relatively uniform, as show in Fig. 2 (c) in cross-section. The spectral emissivity at room temperature of the photonic crystal before and after filling was obtained by measuring near-normal incidence reflectance and reflectance at 45° incidence using a Cary 5000 spectrophotometer. The same reflectance measurement was also performed after annealing the photonic crystal for 24 hours at 1000°C in a quartz tube furnace in vacuum (5×10^{-6} Torr, heating and cooling rate of 10°C/minute). A 20 nm conformal coating

was deposited on the non-filled photonic crystal to prevent degradation of the optical properties during anneal. In simulations, the unfilled photonic crystal was found to have effective (least squares fit of sigmoid with 1200°C blackbody weighting) hemispherical in-band emissivity of 0.594 and out-of-band emissivity of 0.168 whereas a photonic crystal with HfO_2 filled cavities would have an in-band emissivity of 0.922 and out-of-band emissivity of 0.154, corresponding to a 55% increase in in-band emissivity without a change in out-of-band emissivity [13]. Thus, in an unfilled photonic crystal, the emissivity at 45° (approximately the hemispherically averaged emissivity) is about half of that at normal incidence, but in a filled photonic crystal the two emissivities are about equal. As shown in Fig. 3 (b), the peak normal in-band emittance of our fabricated filled photonic crystal was 0.9, and the 45° peak in-band emittance was 0.8, while the out-of-band emittance approached the emissivity of flat Ta. A sharp cutoff was observed in the desired wavelength range. The optical properties remained remarkably stable even after annealing. The measured emittance of the photonic crystal matched the simulated emittance relatively well, as shown in Fig. 3. This work demonstrates that dielectric filling improved the hemispherical performance of our 2D photonic crystal selective emitter without sacrificing stability or ease of fabrication allowing for further increase in TPV system efficiencies.

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