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# Passive Radiative Cooling of Solar Cells by Low-Cost and Scalable Metamaterials: Physical Simulation and Efficiency Limits

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## ABSTRACT

Radiative cooling is an attractive concept for future sustainable energy strategies, as it might enable passive cooling of buildings and photovoltaic systems, hence facilitating energy savings by boosting performance and lifespan. The key idea is the adoption of materials that strongly emit thermal radiation in the atmosphere transparency window (wavelengths between 8 and 13 μm) as cooling layers. Significant progress in the field of metamaterials has enabled the realization of dielectric photonic structures with properties matching radiative cooling requirements and capable of going below ambient temperature. However, these structures are rather expensive and appear unsuitable for today's large-scale manufacturing. In the present work, we have studied radiative cooling applied to Shockley-Queisser solar cells by exploring alternative materials, namely cementitious phases, which exhibit the required properties while being low-cost and scalable. We have determined their emission behavior by electromagnetic simulations and estimated the corresponding solar cell operating temperature by means of a detailed-balance model. The results have been benchmarked against the current state-of-the-art and hint at the possible realization of a new class of radiative coolers based on cheap and scalable cementitious materials.

**Keywords:** radiative cooling, solar cells, metamaterials, detailed balance, electromagnetic simulations, cements, concrete

## 1. INTRODUCTION

In the past few decades, growing concerns over global warming have prompted scientists to strive for more efficient renewable energy sources. Photovoltaic (PV) systems converting sunlight into electricity have experienced steady performance improvement and cost reduction and gained a prominent role in the market of sustainable energy production systems.

Temperature is one of the most critical environmental factors for the operation of a solar cell (SC).[1] Indeed, the power conversion efficiency decreases with temperature, which can be observed, for example, by calculating the efficiency limit of a single-gap cell according to the model from Shockley and Queisser (SQ).[2] Moreover, a high operating temperature has a negative impact on the reliability of the PV system and reduces its lifespan.[3]

Unfortunately, photovoltaic systems usually operate at rather high temperatures because of the exposure to the Sun; this makes cooling concepts particularly important. Accordingly, the scientific community has provided several thermal-management options over the years.[4] However, many of these technologies require an energy supply to operate (active cooling), hence reducing the actual gain in energy yield. An alternative strategy is based on cooling mechanisms capable of removing heat from a body passively, *e.g.*, radiative cooling.

Radiative cooling is the ability of a body to lower its own temperature by emitting thermal radiation.[5] Bodies emitting radiation within the so-called atmospheric window (AW) between 8 μm and 13 μm wavelengths, where the atmosphere is

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electromagnetically transparent, gain direct access to the cold (approximately 3 K) outer space and can lower their temperature even below the ambient one.[6] This is not possible for bodies emitting radiation outside the AW, since the atmosphere acts as an opaque shield in this spectral region, which sends the radiation back to its source. By thermally coupling such bodies to a photovoltaic device, it should be possible to lower the operating temperature of the latter passively.

Although bodies emitting in the atmospheric window have been realized before,[7] radiative cooling is recently experiencing a strong comeback because of the first experimental realization of passive cooling below ambient temperature with a thin multilayer planar metamaterial made of Ag, Ti, $SiO_2$, and $HfO_2$.[8] Over the following years, other planar and/or patterned metamaterials have been proposed for radiative cooling of buildings and/or photovoltaics on the basis of simulations and/or experiments.[9,10,11,12]

Although these works have provided a proof of principle, they rely on complex and expensive metamaterials, that usually contain rather costly and scarce constituents (e.g., Ag, $HfO_2$), and require fabrication processes hardly compatible with today's large-scale manufacturing. To break this deadlock, we are working together with several partners on an EU-funded project, named MIRACLE, which aims at providing cheap and scalable materials for radiative cooling of buildings and photovoltaic systems.[13,14] The revolutionary idea of MIRACLE is to use concrete as the ultimate "metamaterial" for radiative cooling applications, which would provide a working technology that is cheap and scalable. This at-first-sight crazy idea relies on the complex nature of concrete, which consists of a porous cement matrix filled with different kinds of aggregates. Both the microstructure and the chemistry can be modified with many degrees of freedom, de facto rendering concrete a cheap and tunable metamaterial. The success of the project could lead to significant energy savings for the cooling of buildings. Furthermore, the possibility of integrating this engineered concrete into a photovoltaic panel could lead to the realization of a cheap and scalable passive cooling concept for photovoltaic systems.

In this study, we want to provide a preliminary assessment of the potential of concrete for radiative cooling of photovoltaic devices, by studying the impact of radiative coolers made of the main components of ordinary Portland cements (OPC) on the operating temperature of solar cells.

OPCs are usually formed by mixing a clinker powder with water. The clinker components (clinker phases) produce different kinds of hydrates (hydration phases) by reacting with water, leading to the formation of a gluing gel. As this hydration process goes on, the cement hardens. The main clinker phase of OPC is tricalcium silicate (chemical formula: $Ca_3SiO_5$ - cement chemistry notation: $C_3S$), while the main hydration phases are calcium hydroxide (chemical formula: $Ca(OH)_2$; cement chemistry notation: CH) and calcium silicate hydrate (chemical formula: 0.6-2.0 $CaO \cdot SiO_2 \cdot$ 0.9-2.5 $H_2O$; cement chemistry notation: CSH). The curious reader can refer to the relevant literature for further details on the cement materials properties, which are beyond the scope of this proceeding.[15]

We have investigated the electromagnetic emission properties of these three phases and estimated their capabilities in terms of radiative cooling of photovoltaic devices. We have assumed a planar geometry without limiting ourselves in terms of thickness, since one could envision both the integration of a photovoltaic panel into a building rooftop made of the radiative-cooling concrete, or the "painting" of the latter or of the cement phases (without necessarily using them to make concrete blocks) on the photovoltaic panel backside. Finally, we have compared their performance to the theoretical radiative emitter proposed in [16] and to different metamaterials reported in the literature.

## 2. METHODOLOGY

The model system used for our assessment consists of a planar stack of a perfect mirror, a radiative cooler, and a solar cell, with the latter facing the Sun, as studied by Safi and Munday.[16] The radiative cooler and the solar cell are assumed to be decoupled electromagnetically, with the former operating in the IR spectral range and the latter operating in the UV-visible spectral range, but perfectly coupled thermally, *i.e.*, at the same operating temperature $T$. These conditions correspond to ideal design targets and let us treat the radiative cooler and the solar cell as a single body at temperature $T$, whose electromagnetic properties are given by the former in the IR spectral range and by the latter in the UV-visible one. The perfect mirror ensures that all the energy exchanges between the system and the environment take place on the same top surface, enabling us to conveniently describe these processes in terms of power per unit area. Since the atmosphere and the Sun emit radiation in the IR and UV-visible spectral ranges, respectively, the former is assumed to interact only with the radiative cooler and the latter only with the solar cell.

The operating temperature $T$ of the system is determined by solving the equation $P_{\text{net}}(T) = 0$ with respect to $T$, where $P_{\text{net}}$ is the device net power per unit area, which equals zero in the steady state. $P_{\text{net}}$ is given by

$$P_{\text{net}}(T; T_0, E_g, h_c) = P_{\text{rad,cell}}(T; E_g) + P_{\text{rad,cool}}(T; E_g) + P_{\text{elec,cell}}(T; E_g) + P_{\text{con}}(T; T_0, h_c) \\ -P_{\text{abs,sun}}(E_g) - P_{\text{abs,atm}}(T; T_0) \quad (1)$$

according to the model proposed by Perrakis et al.[17] The right-hand-side of Equation 1 contains terms associated to the channels through which Sun, atmosphere, solar cell, radiative cooler, and external load exchange energy, as depicted in Figure 1, by radiation emission (rad) and absorption (abs), thermal conduction/convection (con) or delivery of electricity (elec).

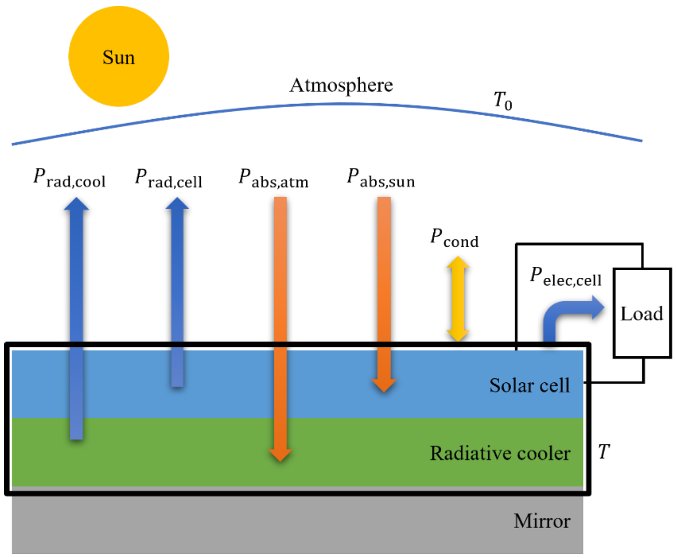


Figure 1. Model system and energy exchange terms used for assessing the potential of the materials investigated for radiative cooling of photovoltaic systems.

Within a spherical coordinate system whose origin is on the top surface of the photovoltaic system and whose zenith angle $\theta$ is measured with respect to the direction normal to the system planar structure, these terms can be calculated as follow for a system with azimuthal invariance, by keeping in mind that absorptivity and emissivity are equivalent by virtue of Kirchhoff's law for thermal radiation:

$$P_{\text{rad,cool}}(T) = 2\pi \int_{\theta=0}^{\pi/2} \cos\theta \ \sin\theta \ \int_{\lambda=0}^{+\infty} \epsilon_{\text{cool}}(\lambda;\theta)\ I_{\text{BB}}(\lambda;T)\ \mathrm{d}\lambda\ \mathrm{d}\theta \quad (2a)$$

$$P_{\text{rad,cell}}(T; E_g) = \pi \int_{E_g}^{+\infty} E\, n\left(E; T, V_{\text{MPP}}(T; E_g)\right) \mathrm{d}E \quad (2b)$$

$$P_{\text{elec,cell}}(T; E_g) = J_{\text{MPP}}(T; E_g)\ V_{\text{MPP}}(T; E_g) \quad (2c)$$

$$P_{\text{abs,atm}}(T; T_0) = 2\pi \int_{\theta=0}^{\pi/2} \cos\theta \ \sin\theta \ \int_{\lambda=0}^{+\infty} \epsilon_{\text{cool}}(\lambda;\theta)\ \epsilon_{\text{atm}}(\lambda;\theta)\ I_{\text{BB}}(\lambda;T_0)\ \mathrm{d}\lambda\ \mathrm{d}\theta \quad (2d)$$

$$P_{\text{abs,sun}}(E_g) = \int_0^{hc/E_g} s_{\text{AM1.5G}}(\lambda)\ \mathrm{d}\lambda \quad (2e)$$

$$P_{\text{con}}(T; T_0, h_c) = h_c\ (T - T_0) \quad (2f)$$

where $h$ and $c$ are Planck's constant and speed of light in vacuum, $\lambda$ is the radiation wavelength, $E$ is the photon energy, $\epsilon_{\text{cool}}(\lambda;\theta)$ is the radiative cooler spectral directional emissivity, $\epsilon_{\text{atm}}(\lambda;\theta)$ is the atmosphere spectral directional emissivity, $E_g$ is the solar cell semiconductor bandgap, $J_{\text{MPP}}$ and $V_{MPP}$ are the solar cell maximum-power-point (MPP) electrical current density and voltage determined according to the SQ model, $h_c$ is the thermal conduction/convection coefficient (set to 10.6 W m$^{-2}$ K$^{-1}$ to represent the case of average winds[17]), $T_0$ is the ambient temperature (set to 293.15 K), $s_{\text{AM1.5G}}(\lambda)$ is the Sun spectral irradiance according to the standard air-mass 1.5 global spectrum,[18] $n(E; T, V)$ is the spectral directional photon flux emitted by a black-body at temperature $T$ under a bias voltage $V$ according to the generalized Planck law,[19] and $I_{\text{BB}}(\lambda; T)$ is the spectral radiance of a black-body at temperature $T$ according to Planck's law.[20]

The spectral directional emissivity of the atmosphere has been determined by calculating the spectral directional transmittivity at normal incidence $\tau_{\text{atm}}(\lambda)$ with the software package LOWTRAN[21] and by applying the formula[22]

$$\epsilon_{\text{atm}}(\lambda; \theta) = 1 - \tau_{\text{atm}}(\lambda)^{1/\cos\theta} \tag{3}$$

On the other hand, the spectral emissivity of the radiative cooler has been calculated from the complex refractive index of the constituent materials by applying the transfer-matrix method (TMM).[23,24]

The complex refractive index of the OPC phases investigated has been determined from their complex dielectric function, which has been calculated by the software package General Lattice Utility Program (GULP)[25] with the polarizable force-field scheme CLAYFF,[26] which is well-established for cementitious phases.[27] The complex refractive index of the films forming the multilayer planar metamaterials investigated has been taken from the online database RefractiveIndex.INFO.[28]

Both p and s polarization of the incident radiation have been considered and the emissivity results have been averaged before being inserted in the energy balance model defined by Equation 1.

## 3. DATA

We have compared the PV radiative cooling performance of the studied ordinary Portland cement phases to the theoretical emitter proposed by Safi and Munday and to several metamaterials reported in literature. The radiative cooler of Safi and Munday has unit emission in the spectral range between 8 µm and 26 µm and zero emission elsewhere.[16] Concerning metamaterials, we have considered the ones proposed by Raman et al., which is formed by a stacking of Ag, Ti, $SiO_2$ and $HfO_2$,[8] and by Kecebas et al., which consist of a stack of Ag, $TiO_2$ and $SiO_2$ or a stack of Ag, $TiO_2$, $SiO_2$ and $Al_2O_3$.[11] We have obtained the best performance for the configuration Ag(50 nm) / $TiO_2$(20 nm):$SiO_2$(20 nm) × 2 / $TiO_2$(200 nm):$SiO_2$(200 nm):Al2O3(200 nm) × 3; hence, we use this configuration as representative for all metamaterials studied to benchmark the OPC phases investigated. We have considered radiation incidence angles from 0° up to 90°. The calculated spectral directional emissivity is reported in Figure 2.

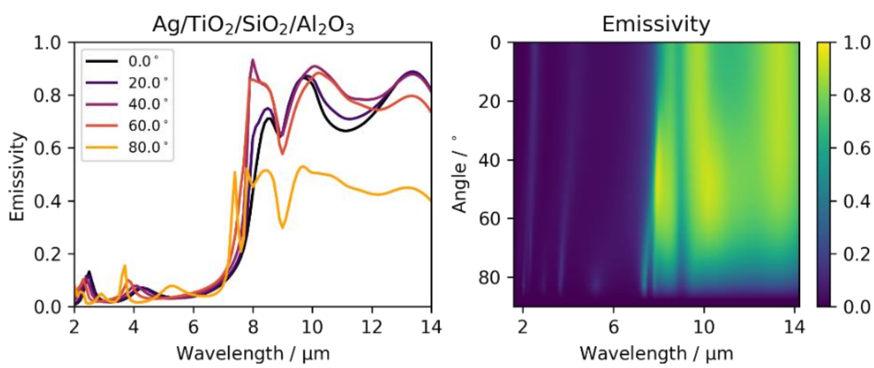


Figure 2. Spectral directional emissivity of the radiative-cooling metamaterial proposed by Kecebas et al. and formed by the stack Ag(50 nm) / $TiO_2$(20 nm):$SiO_2$(20 nm) × 2 / $TiO_2$(200 nm):$SiO_2$(200 nm):Al2O3(200 nm) × 3.

For the OPC phases $C_3S$, CH and CSH, we have calculated the spectral directional emissivity of single layers with thicknesses ranging from 1 µm to 10 cm, to avoid limiting ourselves to specific configurations and technologies at this explorative and speculative stage, as justified in the Introduction section. 1 mm thickness has provided the best results; hence, the corresponding spectral directional emissivity is reported in Figure 3 for all phases.

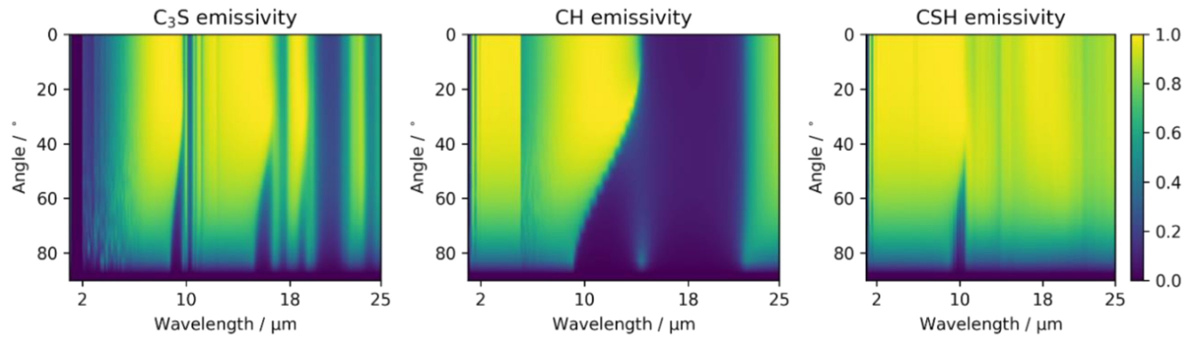


Figure 3. Spectral directional emissivity of 1mm thick layers of the main clinker ($C_3S$) and hydration (CH, CSH) phases of ordinary Portland cements.

Finally, we have compared the operating temperature of a SQ solar cell as function of the semiconductor bandgap, when combined with the theoretical radiative cooler from Safi and Munday, with the $Ag/TiO_2/SiO_2/Al_2O_3$ radiative cooler and with the OPC phases hereby investigated. The results are reported in Figure 4, where also the scenario without cooler is included to assess the cooling effect.

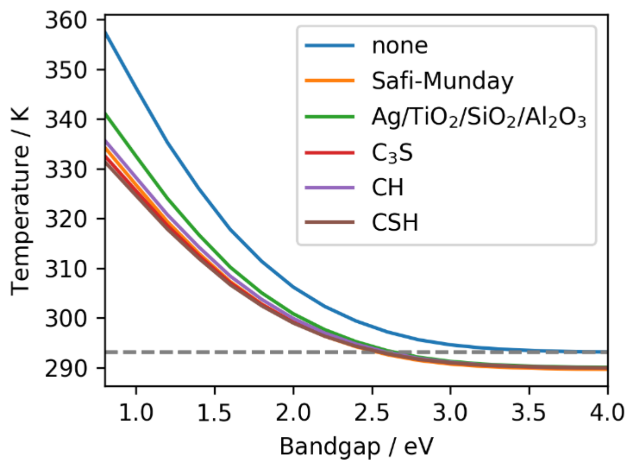


Figure 4. Operating temperature vs semiconductor bandgap of a SQ solar cell without radiative cooler (“none”) or coupled with the radiative coolers “Safi-Munday” and $Ag/TiO_2/SiO_2/Al_2O_3$ or coupled with radiative coolers made of the OPC main phases $C_3S$, CH and CSH. The dashed line represents ambient temperature, set at 293.15 K.

## 4. RESULTS

First and foremost, all simulated materials emit a significant amount of thermal radiation in the atmospheric window and can cool down a SQ solar cell (by up to 25 K) with arbitrary bandgap, according to the model adopted for our assessment.

Concerning the $Ag/TiO_2/SiO_2$ and $Ag/TiO_2/SiO_2/Al_2O_3$ metamaterials, we could generally reproduce the results reported by Kecebas et al. for different kind of configurations.

Concerning concrete, all the OPC phases studied strongly emit thermal radiation within the atmospheric window and beyond, as shown in Figure 3, and are capable of radiatively cooling SQ solar cells quite effectively, as can be seen from Figure 4. This is a remarkable result since it suggests that cheap and scalable cement phases can be potentially employed for radiative cooling of photovoltaic systems. What is more striking, is that their performance approaches the one of the theoretical radiative cooler proposed by Safi and Munday. This can be explained by the broader emission spectral range of the OPC phases, which unlike the theoretical cooler emit also below 8 µm and can take advantage of another atmospheric window at 4 µm. Therefore, the OPC phases seem to be capable of competing if not surpassing the more expensive metamaterials.

However, it is important to remark that this is a preliminary assessment that neglects certain important aspects of concrete technology, such as the porous structure, the presence of many other phases within the material (clinker phases, hydration phases, aggregates), and the surface roughness, which can range from few to tenths of micrometers depending on the polishing,[29] hence being comparable to the wavelengths involved. The complex microstructure and chemical landscape are not necessarily a disadvantage since they can provide more room for tailoring the cement and concrete properties to the application of interest. On the other hand, surface roughness appears problematic, but nothing is going to forbid the use of the cement phases as thin "paintings" for the photovoltaic system bottom side instead of components of an actual concrete wall, which would reduce the surface-morphology related issues.

## 5. CONCLUSIONS

We have provided a preliminary assessment of the potential of the main components of ordinary Portland cements as passive radiative coolers for photovoltaic systems. Based on a detailed-balance model, we have found out that the $C_3S$ clinker phase and the CH and CSH hydration phases exhibit strong emissivity of thermal radiation in the atmospheric transparency window. Accordingly, they appear capable of reducing the operating temperature of a photovoltaic device with arbitrary bandgap by radiative cooling. More strikingly, the cooling performance of these materials seems to compete with more expensive complex metamaterials reported in literature. Further investigations and studies are needed to better assess the feasibility of a radiative cooler made of these OPC phases, in the form of actual concrete or cement, or in other forms (such as "painted" thin films) that are cheap and scalable. For the former, it is important to keep into account all aspects of concrete technology, such as porous microstructure, surface roughness, mechanical stability at thin geometries, complex chemical landscape, and technological challenges in terms of integration into an actual photovoltaic system. However, these first results are very encouraging and call for further research since the realization of photovoltaic systems enhanced by cost-effective and scalable passive cooling would represent a real technological breakthrough.

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