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UNIDIRECTIONAL LUMINESCENT SOLAR CONCENTRATORS

Luminescent solar concentrators have the potential to create low-cost solar energy conversion systems with reasonable conversion efficiency, if one succeeds in sufficiently reducing waveguide losses. Together with non-radiative recombination in the luminophores, escape cone losses of light that exits the waveguide before reaching the solar cells is one of the main loss mechanisms. It has been shown with optical simulations that these losses can be efficiently reduced by creating anisotropic emitting luminophores, which predominantly emit in the plane of the waveguide. So far, these simulations considered only anisotropy in emission. Reciprocity dictates, that such emitters will also demonstrate anisotropic absorption. In the case of aligned, unidirectional emitting structures, which emit the light in only one direction into the waveguide, the anisotropy in absorption can form an additional benefit in the reduction of photon reabsorption events. As the structures have high emissivity in the forward direction and low emissivity in the backward direction, light absorption from the backside will be reduced. This facilitates efficient traveling of photons in the forward direction and reduces waveguide losses. By implementing the effects of anisotropic absorption in a Monte Carlo ray-tracing model (MC model), the effect on luminescent solar concentrator performance is investigated. A large increase in efficiency is found, especially for high geometric gain values, while scattering losses due to the high index structures turn out negligible. This reveals the promising potential of unidirectional emitters for high efficiency, high geometric gain luminescent solar concentrators.

7.1 INTRODUCTION

Luminescent solar concentrators (LSCs) are gaining interest with the growing demand for photovoltaic (PV) systems for the transition to renewable energy.²²⁵ Due to their ability to concentrate light from a large area with cheap glass or polymer sheets to small area solar cells, they can help in overcoming some of the foreseen material scarcity for the production of PV systems.⁴ Furthermore, with their potential for semi-transparent and colorful designs, they are excellent candidates for building integrated PV¹⁸⁵ and other niche applications.²²⁵ In the past decades, large improvements have been made in creating highly efficient luminophores, which are essential for creating efficient LSC devices.²²⁶ In chapter 6, we have shown the great potential of engineering anisotropic emission patterns to reduce the escape cone losses, which up till now form one of the major loss mechanisms in LSCs. We showed that reducing escape cone losses is equally important as reducing the non-radiative recombination in the luminophores. In the analysis, we only focused on the effects of the emission pattern. This was done by implementing anisotropic emission patterns in a detailed Monte Carlo ray-tracing model. However, due to rules of reciprocity, also the absorption pattern will be influenced and become angular dependent and additional scattering might be induced. Given the encouraging results for anisotropic emitters, and especially the unidirectional forward emitting nanocones, a more detailed investigation including all optical effects is needed to reveal the full potential of this concept.

Anisotropic absorption will have both positive and negative consequences for the LSC performance, and the balance between these deserves further investigation. On the one hand, reduced emission into the escape cone will also lower the absorption of incoming sunlight. On the other hand, in the case of the unidirectional, aligned nanocones, reabsorption of photons traveling in the waveguide will be reduced. Again due to reciprocity, high emissivity in the forward direction and low emissivity in the backward direction will result in low absorption from the backside and high absorption from the front side. Light emitted by the luminophores will predominantly travel in the forward direction, and thus experience low absorption probability in the next, aligned nanocone. Any photons emitted in the backward direction will have a large chance of reabsorption, with subsequent re-emission in the forward direction. The path traveled by the photons in the waveguide will no longer be a random walk like it is for isotropic emitting luminophores, but will be a more straight path towards the solar cells. This reduces the total path length, and thereby reduces reabsorption probability even further. Finally, there is the presence of higher index nanophotonic structures in the waveguide, which introduce additional scattering. The amount and direction of this scattering also require further investigation.

In this work, we have implemented anisotropic absorption and additional scattering in the Monte Carlo ray-tracing model. To obtain the best LSC performance with these aspects included in the model, a new particle swarm optimization was run for the design of the unidirectional emitting nanocone. This time the structure was not only optimized for maximizing emission in the forward direction, but also for maximizing sunlight absorption and minimizing scattering. With these changes implemented in the Monte Carlo ray-tracing model, we analyze LSC performance for several design parameters.

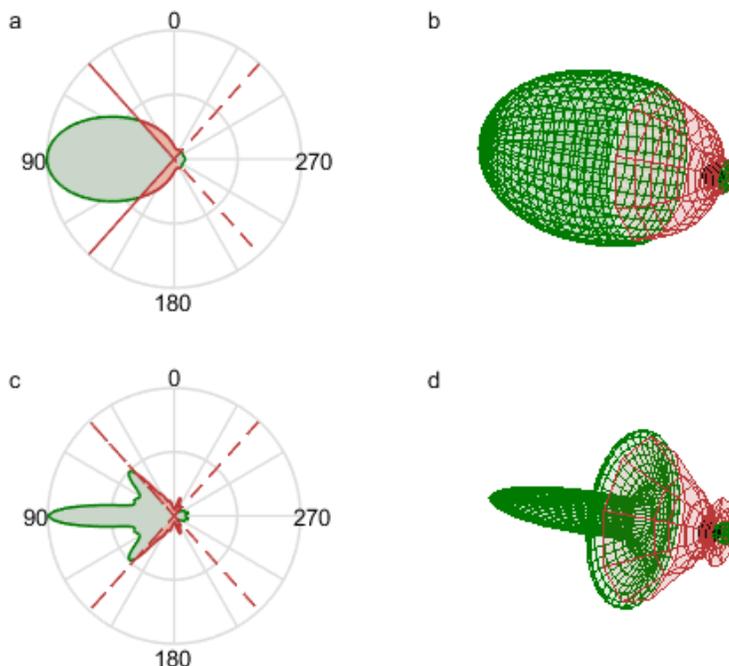


Figure 7.1: Emission and scattering profiles of the forward emitting nanocones. (a) and (b) 2D and 3D polar plots of the emission profile of a luminophore embedded in the nanocone, emitting at 850 nm. 49% of the light is emitted in forward direction and 16.5% is emitted into the escape cone. (c) and (d) 2D and 3D polar plots of the emission profile from an ensemble of 20 aligned but randomly positioned nanocones. Scattering with surrounding nanocones only slightly reduces the forward fraction to 45% and increases the escape cone losses to 17.5%. Parts in green experience total internal reflection and are guided by the waveguide, while parts in red are lost through the escape cone.

7.2 RESULTS AND DISCUSSION

A new particle swarm optimization was used to find a structure with a combination of high forward emission and sunlight absorption and low scattering. In addition to the two radii and length of the cone, also the refractive index was left as an optimization parameter, as this parameter plays an important role in the amount of scattering. The figure of merit was defined as the fraction of forward emission plus the amount of absorption minus the amount of scattering, each normalized to a value close to one, resulting in equal weight in the figure of merit calculation. The forward emission and scattering were calculated at the luminophore emission wavelength of 850 nm and absorption was determined at normal incidence in the wavelength range of 400 to 650 nm. From an initial random particle swarm with an average figure of merit of 0.79, the structures were optimized to an average figure of merit of 1.17. The best performing structure is a cone with end radii of 65 and 325 nm, a length of 234 nm, and a refractive index of 3.3 and has a figure of merit of 1.51. This structure has five times more absorption and two times less scattering than the forward emitter in the previous chapter, which results in ten times lower scattering for the same amount of sunlight absorption. The emission pattern is shown in figure 7.1 a and b.

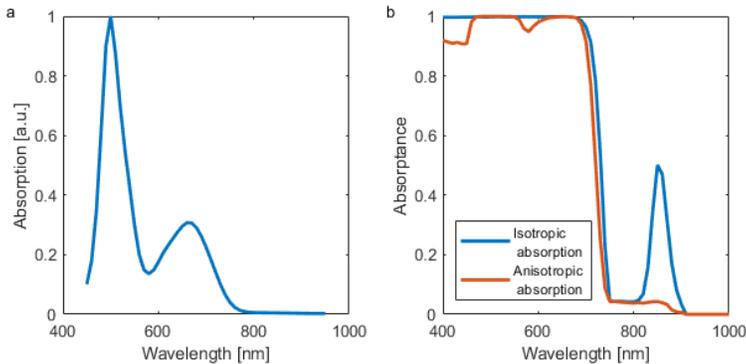


Figure 7.2: Absorption spectra of new optimized forward emitting nanocones. (a) Absorption spectrum of only the cone, as obtained from FDTD calculations with fixed imaginary part of the refractive index for all wavelengths. Due to the combined optimization for high absorption up to 700 nm and high emissivity in forward direction at 850 nm, absorption for normal incidence drops above 700 nm. (b) Absorption spectrum of the LSC device as obtained from the MC model, with and without the anisotropic absorption of the nanostructures implemented. Anisotropic absorption slightly reduces sunlight absorption, especially at the emission wavelength of the luminophores.

Scattering due to the nanocones is primarily problematic for light emitted by the luminophores: these are the photons that have to travel along the waveguide towards the PV cells, and thus have many chances of interacting with adjacent nanocones, while ideally they experience minimal disturbance along their way. We thus looked at scattering of forward traveling light, and investigated how the emission profile is altered after interaction with surrounding nanocones. When a plane wave hits the nanocones in forward direction, there is predominantly forward scattering, with a forward fraction of 60%. The performance of the nanocones is significantly better than the scattering behavior from isotropic particles like conventional luminophores, which give a forward fraction of 17%. This indicates that scattering will not significantly decrease LSC performance. When looking at an ensemble of nanocones, we observe only a small decrease in the forward fraction. The angular emission pattern resulting from an ensemble of 20 aligned, but randomly positioned nanocones is shown in figure 7.1c and d, and has a fraction of 45% of light going in forward direction and 17.5% into the escape cone. For random scattering in the waveguide material, there is no decrease in performance for scattering distances down to 2.5 mm, where efficiency starts to drop. This is in agreement with results for isotropic emitters.²²² A complete analysis of the effect of scattering due to the nanocones would require the implementation of the full scattering profile for each possible angle of incidence of the cone. Since most light will be traveling in the forward direction through the waveguide, we believe that the current analysis is sufficient to conclude that increased scattering due to the presence of the nanocones will not significantly degrade performance.

The absorption spectrum for normal incidence purely due to the optical effects of the nanocone is shown in figure 7.2a. This spectrum is generated disregarding the absorption spectrum of the luminophore, by using a fixed absorption coefficient over all wavelengths. Over the visible part of the solar spectrum, for which the absorption was optimized, absorption is high. Above 700 nm it starts to drop, and at the emission wavelength of 850

nm absorptivity and emissivity in normal direction are almost zero, since the emission is guided in the forward direction. By implementing this nanocone absorption spectrum in the MC model, where it is combined with the absorption spectrum of the luminophores, the absorption spectrum of the LSC device can be determined. For approximately the same optical density over the visible spectrum, implementing the anisotropic absorption properties leads to a small decrease in sunlight absorption, as shown in figure 7.2b, especially at the emission wavelength of the luminophores. However, as we will see in the following, this lower absorption can be compensated by a higher luminophore loading, as reabsorption is reduced with the anisotropic absorption implemented.

With the absorption spectrum and angular profile implemented in the model, LSC performance was investigated for several design parameters. By comparing the nanocone design of the previous work, with ten times more scattering, to the new optimized design, we find an absolute increase of 2% in efficiency for the same optical density of 1, a geometric gain of 10, and photoluminescent quantum yield (PLQY) of 95%, revealing the importance of the three-fold optimization. Figure 7.3a shows the efficiency of the devices with the new cone design for varying optical density (OD), PLQY of 80% and 95%, and a geometric gain (GG) of 10, and shows the effect of implementing the anisotropic absorption properties. Performance is significantly increased by the implementation of anisotropy in absorption. For a PLQY of 95%, the maximum efficiency goes up from 11% to over 16%. While for isotropic absorption the efficiency drops for optical densities above 0.5 due to increased reabsorption losses, the anisotropic absorber peaks at an optical density of 1 and efficiency barely drops for higher OD values. These trends hold independent of luminophore PLQY. For increasing geometric gain the benefit becomes even more pronounced, as visible in figure 7.3b. The isotropic absorber shows a rapid drop in efficiency with increasing GG, but with anisotropic absorption, photons travel over longer distances with lower losses, and efficiency stays above 10% up to a GG of 75. One could think that solar cells on only one out of four LSC waveguide edges are required with this unidirectional emitter design, resulting in a four-times higher geometric gain. However, simulations show that it is still beneficial to have them on all four sides. With 50% of light emitted in the forward direction, the other half is still partly guided in other directions, resulting in the efficiency being halved with only one PV cell on the forward edge. As visible in figure 7.3b, the efficiency goes down by only a few percent when the collection area is increased by factor four, while keeping PV cells on all edges, which is thus the preferred way to increase GG. The low drop in efficiency with increasing geometric gain opens up possibilities for high efficiency, high geometric gain LSCs.

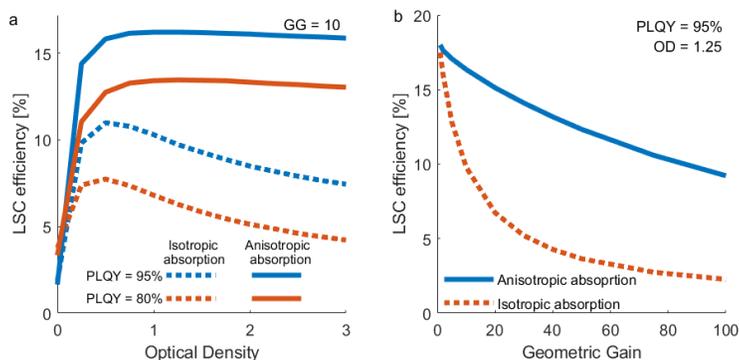


Figure 7.3: LSC performance with forward emitting nanocones, comparing the model with and without anisotropic absorption included. (a) LSC efficiency for varying optical density, for geometric gain of 10 and 20. Due to reduced reabsorption probability with anisotropic absorption included, higher optical densities can be used, and efficiency is significantly enhanced. The penalty for increased geometric gain is lower. This becomes more visible in (b), with efficiency as a function of geometric gain. With isotropic absorbing structures, efficiency drops rapidly due to the large losses associated with reabsorption. With anisotropic absorption, these losses are reduced and efficiency stays above 10% up to a geometric gain of 75.

7.3 CONCLUSIONS AND OUTLOOK

From this analysis, we can conclude that the effects of anisotropic absorption profiles, which are inherently related to anisotropic emission profiles, have a significant effect on LSC performance. Although the forward emitting nanocones cause a small reduction in sunlight absorption and increase scattering, the net result is a large efficiency increase due to reduced reabsorption probability and efficient paths towards the photovoltaic cells. The reduction in sunlight absorption can be compensated with higher luminophore loading, without directly being penalized with higher reabsorption losses. Light traveling in the forward direction through the waveguide will predominantly experience forward scattering from the nanocones, which therefore also does not lead to higher losses. To reach this situation, optimization of the nanocones for directional emission, high sunlight absorption, and low scattering is necessary.

In the current optimization, we focused on investigating the physical principles and their effects on performance. Practical fabrication limitations were not considered, to first see the maximum gain that can be obtained with this concept. As the improvements found are large, also with practical limitations the system has great potential. The next step would be a theoretical design and optimization of unidirectional emitting structures that can be fabricated for experimental verification and subsequent experimental realization.

