

Modeling and Fabrication of Randomly Close Packed Nanostructures using Non-Monodispersed Colloidal Particles

Timothy Chen, I-Te Chen, Chih-Hao Chang*

Department of Mechanical and Aerospace Engineering, North Carolina State University
Raleigh, NC 27695, USA

Light-emitting diodes (LEDs) are considered a viable option for sustainable lighting, but they provide poor emission efficiency of around 20% due to light trapping in the high index layer [1-3]. Integrating nanostructures, such as random surface roughening [2] and photonic crystals [3], can potentially enhance light extraction and increase the efficiency of these devices [1-3]. However, the use of random structure is difficult to control and their geometry cannot be readily designed. On the other hand, the use of periodic photonic crystal can lead to wavelength and angle-sensitive behavior, and can have high fabrication cost. Therefore, there still lacks a scalable fabrication method that allows design of broad bandwidth and wide-angle light extraction, which is desirable for next-generation LEDs for solid state lighting and full-color flat panel displays.

In this work, we investigate a novel approach for modeling and manufacturing of light-extraction nanostructures from high-index medium. This method is based on the co-assembly of non-monodispersed nanoparticles, which can lead to randomly close packed assembly. The first step is to examine the co-assembly process between two distinct diameters at various mixture ratios and distributions, as shown in Figure 1. Here the assembly parameters are 1000 nm particles for monodispersed and 1-to-1 ratio of 500 and 350 nm particles for co-assembly. In this algorithm, a specified number of particles are each assigned a diameter size that can vary by 10% and is based on the mixture ratio. The first particle is always constrained with its center at the origin, and the second particle is set adjacent to the first at a random angle. The positions of the remaining particles are generated by running a loop in MATLAB, each using two adjacent or nearby particles as references. The spatial-frequency spectra of the assembly can then be analyzed based on their intensity distribution of the rings using FFT, as shown in Figure 2. This MATLAB code was run for various mixture ratios among different particle size pairings in order to identify which assembly conditions provide broad spectra band and angle uniformity. Based on the result of the simulated assembly and spectra, randomly close packed nanoparticles were fabricated using the transfer coating method. This is depicted in the top-view SEM images shown in Figure 3, where the assembly conditions are 1000 nm particles for monodispersed and 1-to-1 ratio of 500 and 350 nm particles for co-assembly. The spectrum for the images are also shown in the inset diagram, which closely resemble the spectra for the simulated assembly.

These results demonstrate the proof-of-concept that randomized close-packing can be patterned using colloidal particles with different sizes. The MATLAB script was shown to accurately predict the spectra of the random assembly, and we will present the quantitative analysis of the angular uniformity and spectra bandwidth between the theoretical model and experimental data to validate the constructed assembly model. A parametric study will also be conducted to determine the optimal conditions for using this fabrication method and the accuracy of the prediction model. The script will also be further optimized for predictive design of broadband, wide-angle light extraction.

*tschen@ncsu.edu

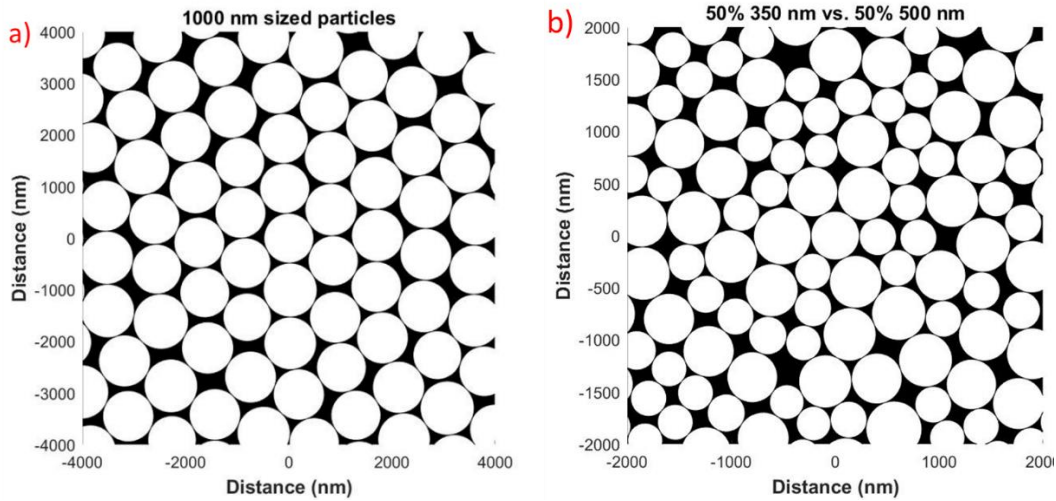


Figure 1. Simulation of randomly close packed particles. The assembly parameters are (a) monodispersed 1000 nm particles and (b) 1-to-1 ratio of 500 and 350 nm particles for co-assembly.

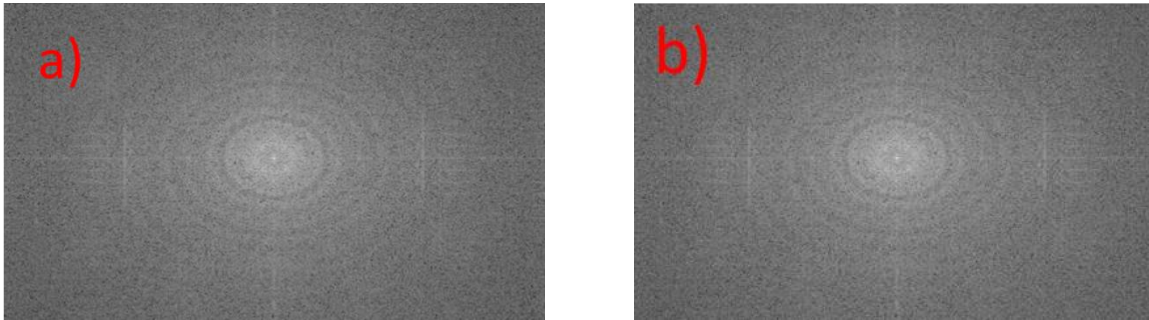


Figure 2. FFT image analysis of images corresponding to Figure 1.

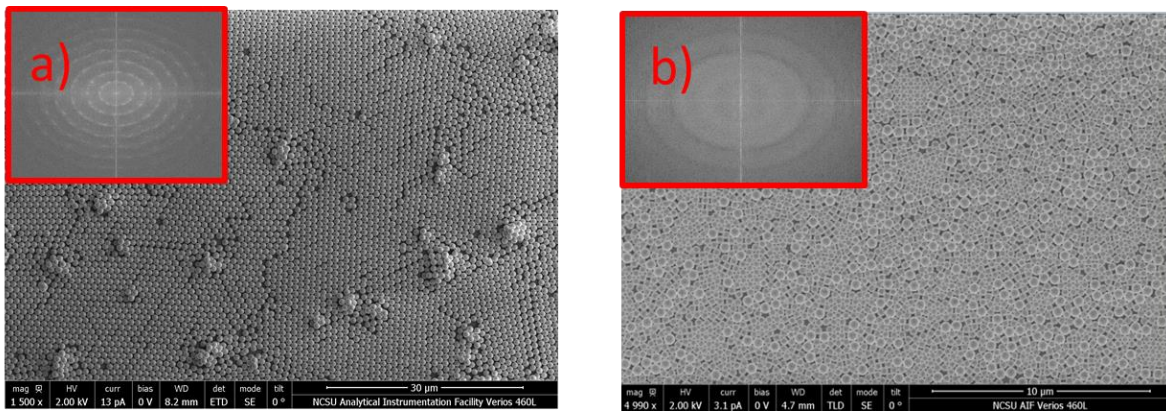


Figure 3. SEM Image of patterned particles. The assembly parameters are (a) monodispersed 1000 nm particles and (b) 1-to-1 ratio of 500 and 350 nm particles for co-assembly.

References:

- [1] Bi, Yan-Gang, et al. *Advanced Materials* 25.48 (2013): 6969-6974.
- [2] Fujii, T., et al. *Applied physics letters* 84.6 (2004): 855-857.
- [3] J.-G. Kim, et al. *Opt. Express, OE*, vol. 23, no. 17, pp. 22730–22739, Aug. 2015.