## Fabrication of Multilayer Complex Nanomesh Patterns

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One of the most interesting and applicable three-dimensional BCP structures is the nanomesh formed by bilayer or multilayer stacks of orthogonal line patterns. Nanomeshes can be useful in several technologies including photonic materials, graphene nanomesh devices or next-generation integrated-circuit architecture. Fabrication of nanomesh structures is restricted to only a few methods, such as using high-resolution electron-beam lithography (EBL)<sup>1</sup> or stacking and aligning layers on top of each other<sup>2</sup>. These methods have several limitations such as the throughput of EBL, lack of precise alignment and difficulty in making complex geometries for layer-by-layer methods. Here, we used orthogonal self-assembly to produce complex multi-layer nanomesh structures without requiring alignment or high-resolution lithographic templating. This finding is scientifically important in that it presents a template-based bottom-up route to create complex multilevel nanomesh patterns. We showed BCP grating patterns can be used to orthogonally self-assemble another layer of a BCP grating with a different period. We have successfully fabricated nanomesh patterns in circles, bends, junctions and with multiple local directions<sup>3</sup>.

Figure 1 shows the major steps of the process. Poly(styrene-*b*-dimethylsiloxane) (PS*b*-PDMS) BCP was spincoated and annealed with solvent vapor at room temperature to facilitate the self-assembly of a monolayer of PDMS cylinders. Reactive ion etching (RIE) of the BCP in a CF<sub>4</sub> followed by an O<sub>2</sub> plasma formed parallel oxidized-PDMS (ox-PDMS) cylinders on the surface of the substrate. Then a monolayer of another PS-*b*-PDMS BCP was spincoated, followed by solvent annealing and RIE similar to the first layer to leave a mesh-shaped structure.

Figures 2 and 3 show the experimental scanning-electron micrographs (SEMs) of the described process. Figure 2-a,b shows the bilayer nanomesh results top and bottom layers of 16 kg/mol PS-b-PDMS (SD16) and 45 kg/mol PS-b-PDMS (SD45), respectively. These SEM images show SD16 cylinders orthogonal to the different complex and ordered patterns of SD45 underneath. The dimensions of the nanomesh holes are  $9 \times 21 \text{ nm}^2$ , an aspect ratio more than 2, which is advantageous for some applications like bit-patterned media. This method has the potential to extend to more than two layers. Figure 2-c shows the result of three-layer nanomesh patterns in which three different BCP cylinders (SD45, SD16 and SD10) are perpendicular on top of each other. In order to experimentally highlight the effect of the orthogonal self-assembly on more complex patterns, we used EBL-fabricated templates to direct the self-assembly of the bottom BCP. Figure 3 shows the results of circular and Y-junction nanomesh patterns. Furthermore, we provided a full understanding of the requirements for orthogonal self-assembly based on self-consistent field theory (SCFT) modeling and showed how it can be generalized (Figure 4). As a results, we determined that the chemical functionalization and height of the first selfassembled were the key parameters for achieving orthogonal BCP self-assembly.

<sup>&</sup>lt;sup>1</sup> A. Tavakkoli K. G., et al., Science **336**, 1294 (2012).

<sup>&</sup>lt;sup>2</sup> J. W. Jeong, et al., Nat. Commun. 5, 5387 (2014).

<sup>&</sup>lt;sup>3</sup> A. Tavakkoli K. G., et al., Nature Communications 7, 101518 (2016).



Figure 1: The major steps of nanomesh pattern fabrication by orthogonal self-assembly.



**Figure 2:** SEMs of nanomesh structures via orthogonal self-assembly, a) bilayer complex nanomesh, b) bilayer ordered nanomesh, c) trilayer ordered nanomesh. Scale bars, 100 nm.



Figure 3: SEMs of nanomesh pattern in a) circular and b) Y-junction. Scale bars, 500 nm.



**Figure 4:** SCFT simulation of the self-assembly of a diBCP under confinement. *A* and *B* refer to the minority (PDMS) and majority (PS) blocks, respectively.