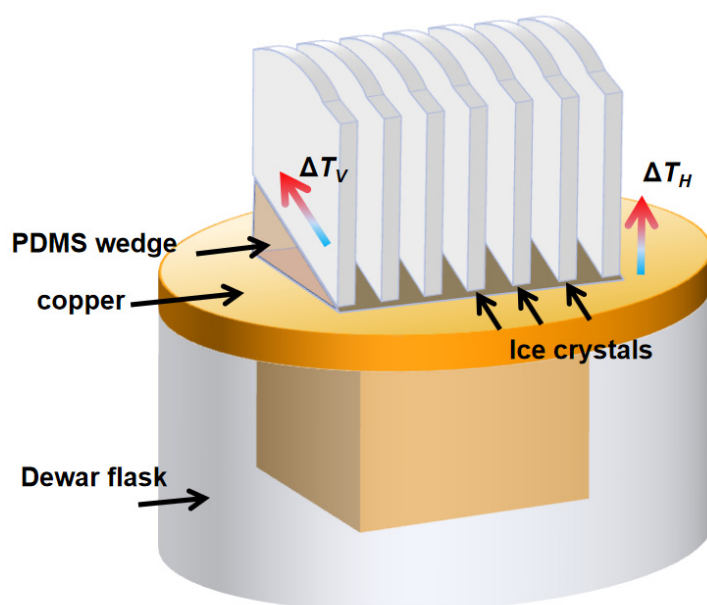
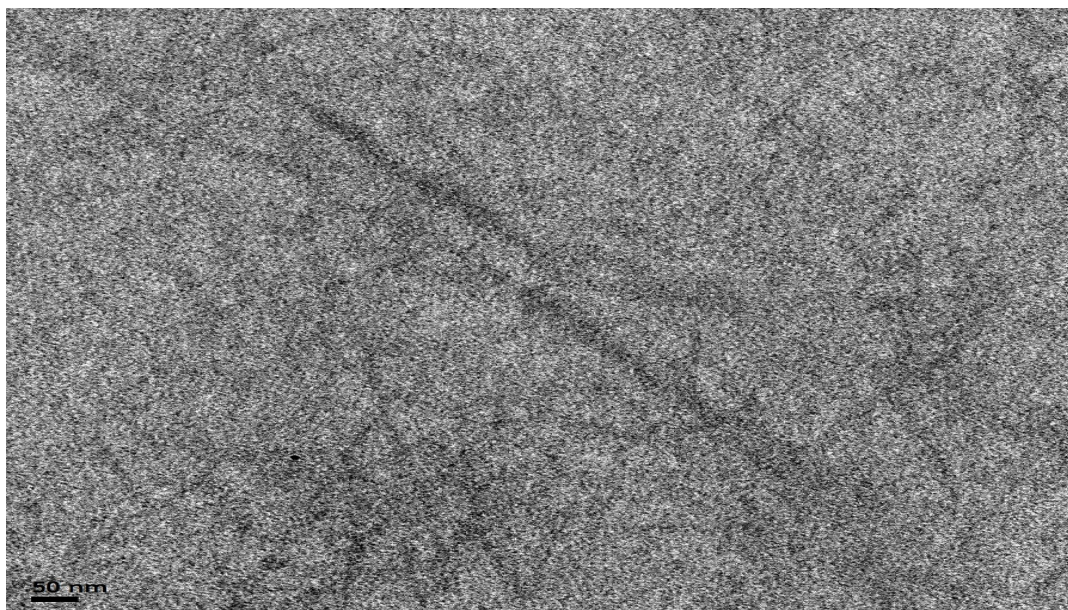


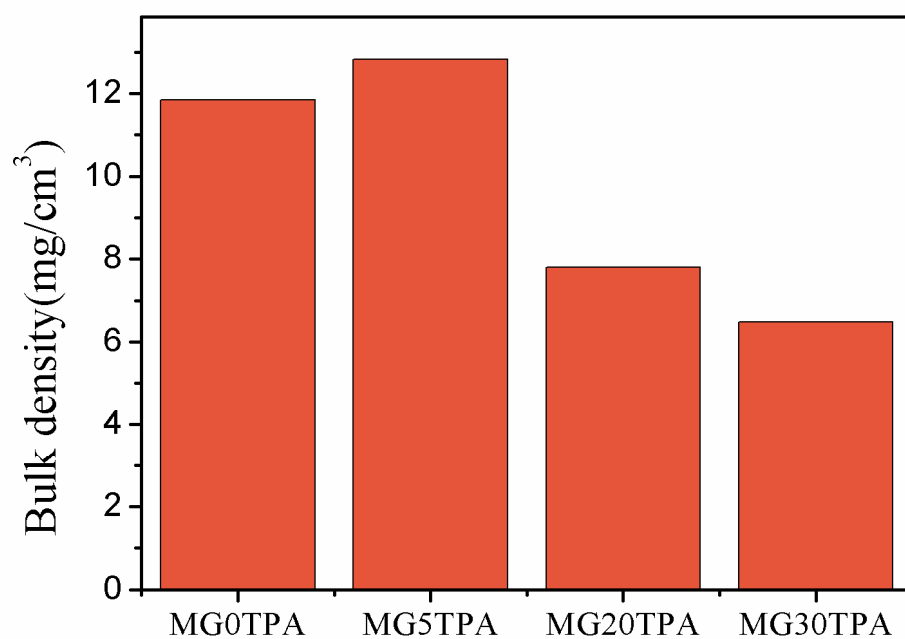
First, the GO/PVA/TOCN suspension was poured into the homemade molds. In order to generate a bidirectional temperature gradient, a PDMS-wedge with low thermal conductivity at a specific Angle ( $15^\circ$ ) is added between the suspension and the cooling copper block. Except for the large temperature gradient in the vertical direction ( $\Delta T_H$ ), there is also a temperature gradient at both ends of the wedge ( $\Delta T_V$ ), due to the existence of the PDMS wedge. The ice crystals grew directionally with gradient along both horizontal direction and vertical direction, which formed parallel and ordered ice columns.



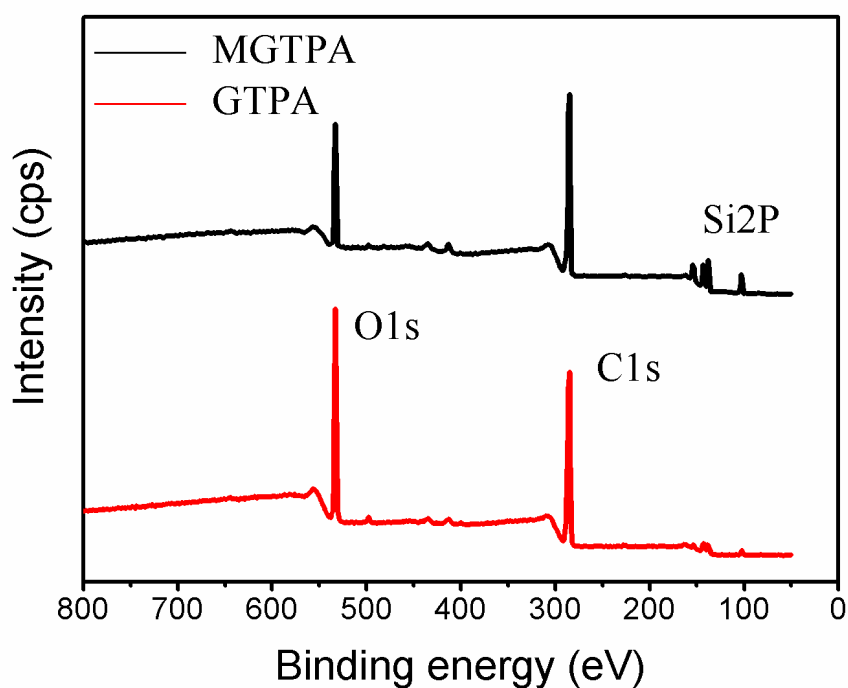
**Figure S1. Schematic illustrations of bidirectional freezing.**



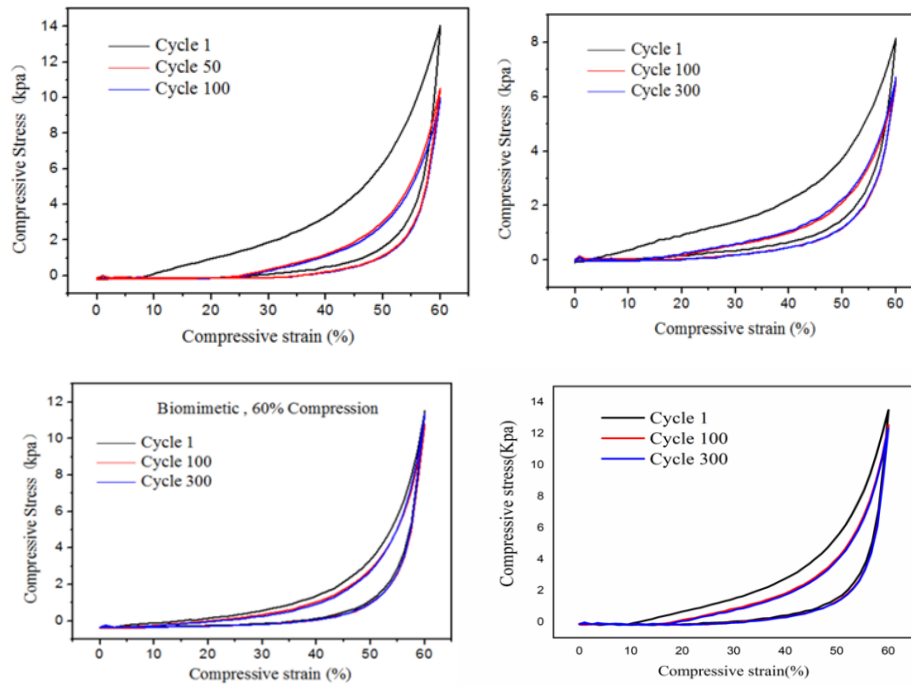
**Figure S2. TEM image of prepared TOCN.**



**Figure S3. The density of the b-MGTPA as a function of initial GO concentration.**



**Figure S4.** XPS survey scans of (a) GTPA and (b) MGTPA. The peak intensity of the C1s peak greatly increased after CVD modification with n-dodecyltriethoxysilane (DDTS), thus indicating grafting of the long carbon chains. Silicon peaks were also detected in MGTPA via XPS.



**Figure S5. Comparison between MGTPA with different GO concentrations in their compression–recovery behaviors (a) MG<sub>0</sub>TPA,(b) MG<sub>5</sub>TPA,(c) MG<sub>20</sub>TPA,(d) MG<sub>30</sub>TPA. When compressed to 60%, the maximum stresses of the MG<sub>0</sub>TPA, MG<sub>5</sub>TPA, MG<sub>20</sub>TPA and MG<sub>30</sub>TPA are 14.06, 8.15, 11.51 and 13.5 kPa, respectively. (c) The aerogel retains 71.6%, 82.5%, 99.3% and 97.4% of its original strength after 100 cycles, respectively.**