Unraveling the Effects of Surface Functional Groups and Assembly Orientations on the Interfacial Mechanics of MXene/Silk Composites

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Figure S1a. Atomic structures of the $Ti_3C_2T_x$ MXene models used in this work, including Ti_3C_2, $Ti_3C_2O_2$, and $Ti_3C_2(OH)_2$.

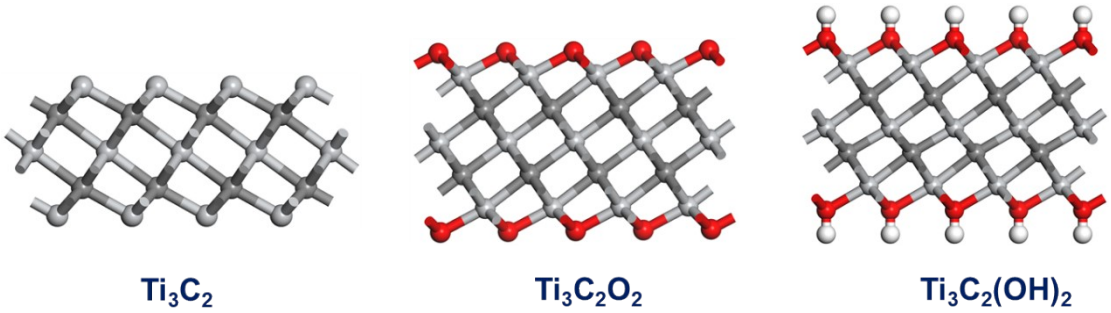

Figure S1b. Three-dimensional representations of the silk fibroin (SF) molecular structure from different viewing orientations.

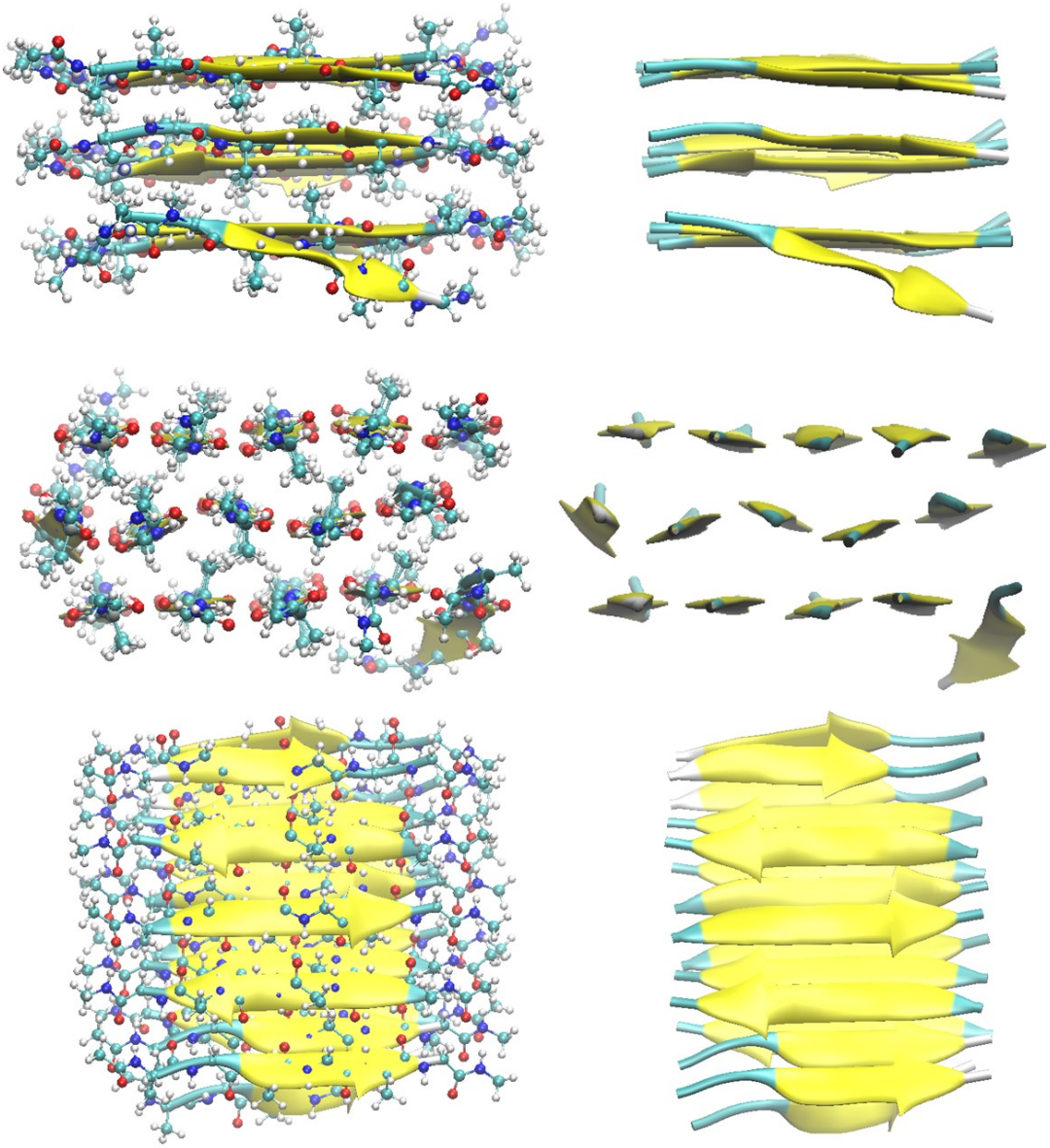

Figure S2a. Layered architecture of MXene (Ti_3C_2)/SF composites with varied chain orientations. The illustrations depict the MXene nanosheets interleaved with SF chains oriented at a) 0° (parallel), b) 45° (diagonal), c) 90° (perpendicular), relative to the MXene surface.

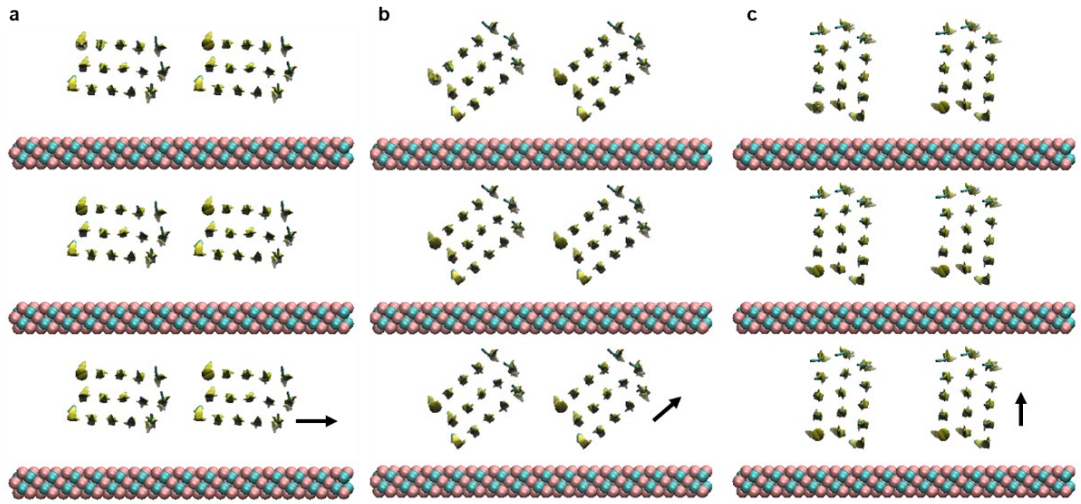

Figure S2b. Layered architecture of MXene ($Ti_3C_2O_2$)/SF composites with varied chain orientations. The illustrations depict the MXene nanosheets interleaved with SF chains oriented at a) 0°, b) 45°, c) 90° relative to the MXene surface.

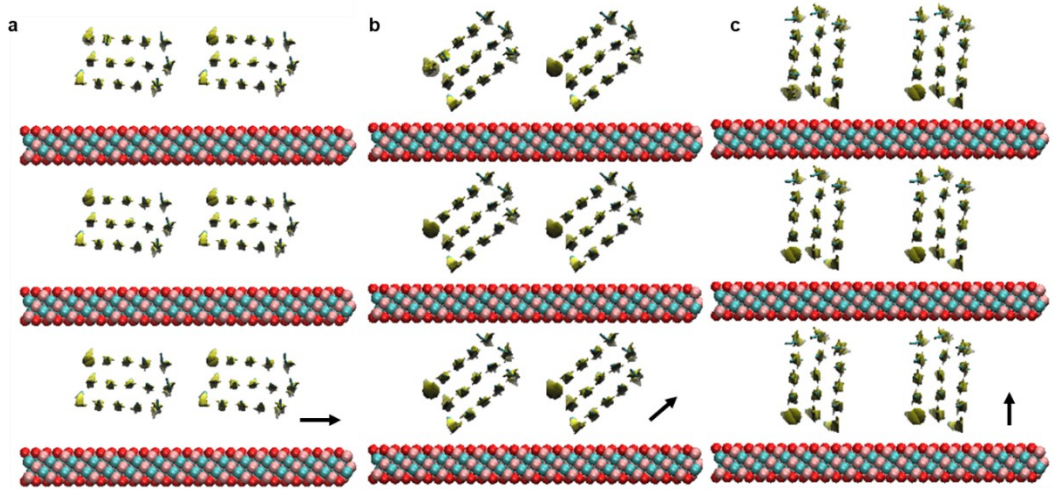

Figure S2c. Layered architecture of MXene ($Ti_3C_2(OH)_2$)/SF composites with varied chain orientations. The illustrations depict the MXene nanosheets interleaved with SF chains oriented at a) 0°, b) 45°, c) 90° relative to the MXene surface.

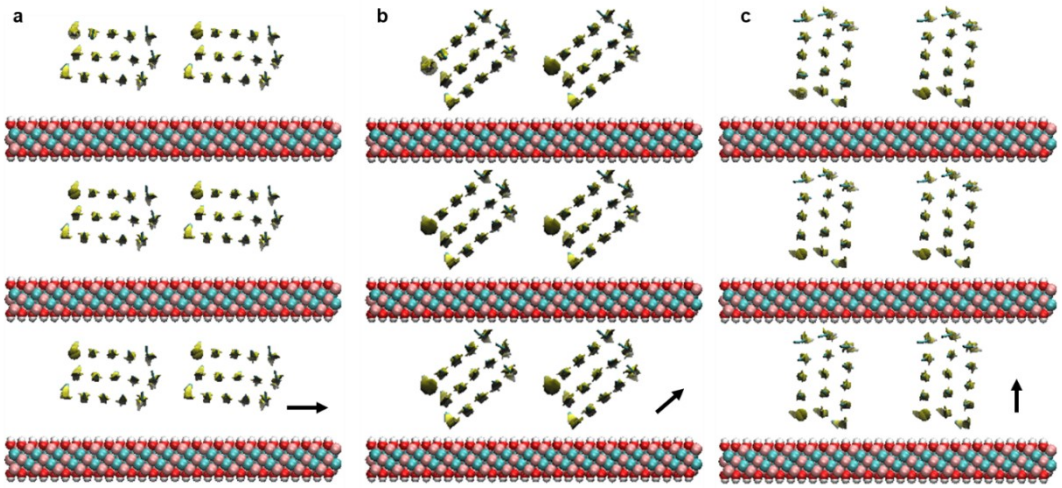

Table S1. The water molecular number in different systems.

Types	Water molecules number
0°_Ti_3C_2/Silk	68257
0°_$Ti_3C_2O_2$/Silk	67310
0°_$Ti_3C_2(OH)_2$/Silk	66496
45°_Ti_3C_2/Silk	82165
45°_$Ti_3C_2O_2$/Silk	80826
45°_$Ti_3C_2(OH)_2$/Silk	81228
90°_Ti_3C_2/Silk	82221
90°_$Ti_3C_2O_2$/Silk	81965
90°_$Ti_3C_2(OH)_2$/Silk	81550

Figure S3. Structural integrity of silk fibroin (SF) during shear deformation. DSSP analysis of the secondary structure composition for SF layers in different MXene/SF composites across 0°, 45° and 90° orientations.

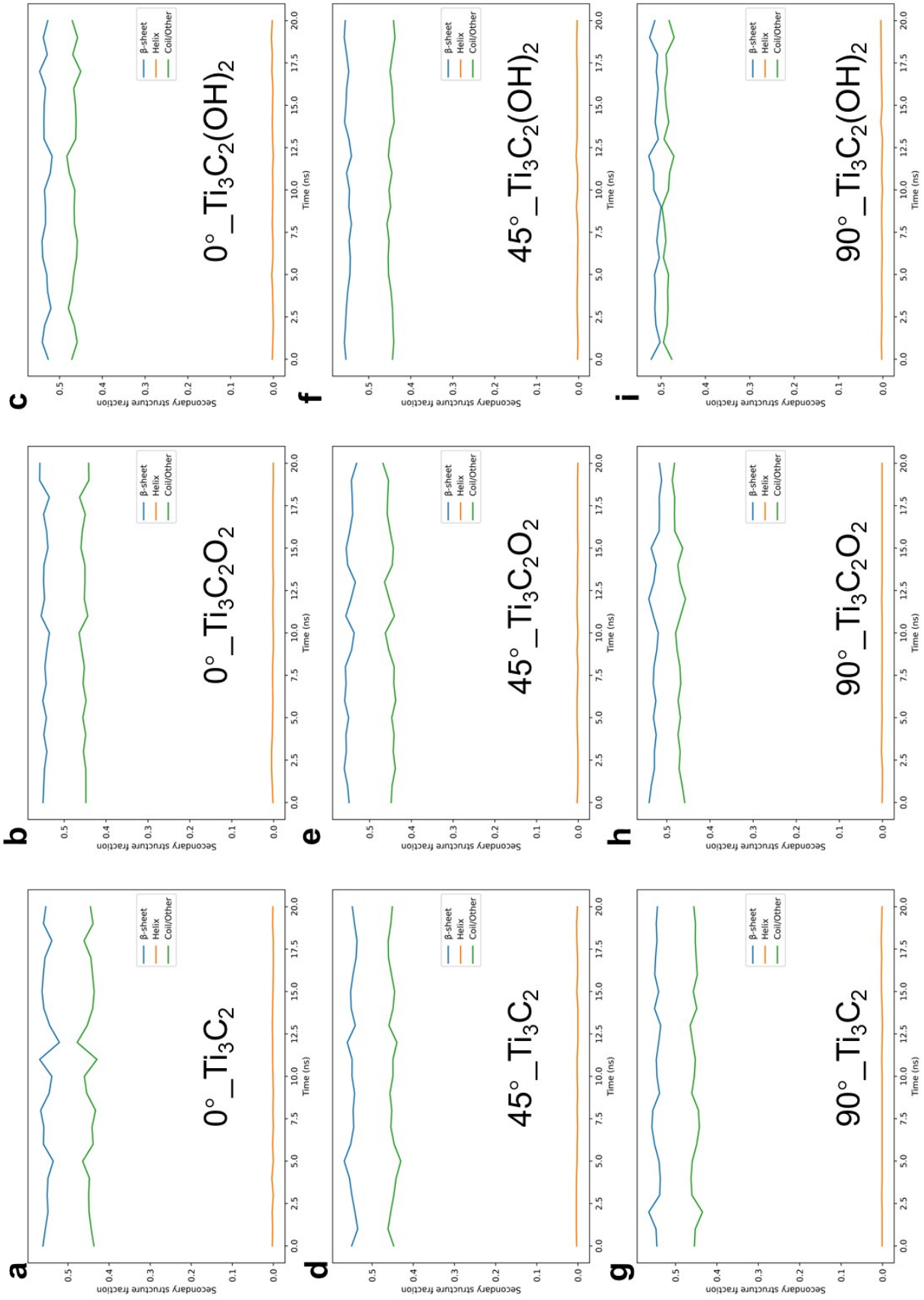

Figure S4. Synergistic Effects of MXene Surface Functionalization and Silk Fibroin Alignment Angle on the Mechanical Performance and its Interfacial Failure Modes.