Description of Additional Supplementary Files

File Name: Supplementary Movie 1

Description: Includes the animation of the CSH-PVA interface during molecular dynamics

simulations.

File Name: Supplementary Movie 2

Description: Includes the animation of the CSH-PVA model during a uniaxial tensile process

of molecular dynamics simulations.

File Name: Supplementary Movie 3

Description: Includes the animation of crack propagation and fracture of cement-hydrogel

under three-point bending stress in the FEM simulation.

File Name: Supplementary Movie 4

Description: Includes the animation of crack propagation and fracture of cement paste

under three-point bending stress in the FEM simulation.