Electronic Supplementary Material (ESI) for Journal of Materials Chemistry B. This journal is © The Royal Society of Chemistry 2021

Supporting Information

Bioprocess-inspired synthesis of multilayered chitosan/CaCO₃ composites with

nacre-like structures and high mechanical properties

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Figure S1. SEM images of CaCO₃ films obtained at different concentration of Mg^{2+} a) 0 mM; b) 5 mM; c) 10 mM; d) 20 mM.



Figure S2. CaCO₃ films obtained at different concentrations of PAA.



Figure S3. a) XRD patterns of CaCO₃ films obtained at different concentrations of PAA. b) XRD patterns of CaCO₃ films obtained at different concentrations of Mg²⁺ with 2010^{-3} wt% PAA.



Figure S4. CaCO₃ films obtained at different concentrations of Mg^{2+} with 2010^{-3} wt%

PAA.

Figure S5. Mechanical properties of $CaCO_3$ films obtained at different concentrations of Mg²⁺ with 2 \circ 10⁻³ wt% PAA. a) Young's modulus-displacement curves. b) Hardness-displacement curves.

Figure S6. Scanning electron microscopy (SEM) images of mineralization directly on (CS/CaCO₃)₁ without spin-coating CS films.

Figure S7. Mineralization at different dripping velocities. a) mineralization for 12 h at a dripping velocity of 4.16 mL h^{-1} . b) mineralization for 72 h at a dripping velocity of 0.69 mL h^{-1} .

Figure S8. Mechanical properties of natural nacre tested by nanoindentation. a) Young's modulus-displacement curves. b) Hardness-displacement curves. c) Loss factor-frequency scatter plots. d) Storage modulus-frequency scatter plots. e) Loss modulus-frequency scatter plots.

Figure S9. TGA data for the synthetic (CS/CaCO₃)₃ and chitosan.

Figure S10. Load-displacement curves for (CS/CaCO₃)₃.