

## Supporting Information

### Title

### Mineralization of Bouligand-structured collagen matrices derived from fish scales with enhanced mechanical properties and biocompatibility

Author(s), and Corresponding Author(s)\*

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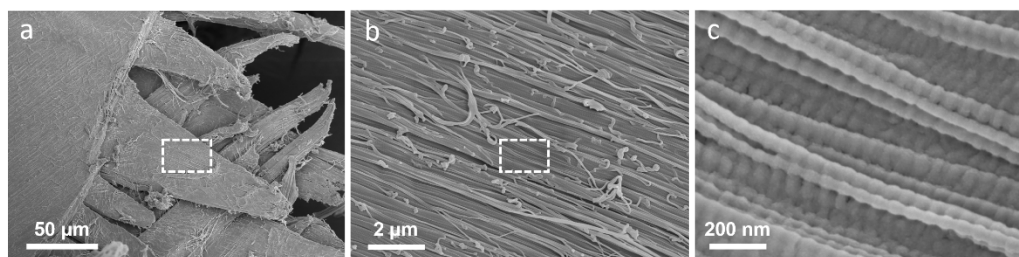


Fig. S1. (a) SEM image of the fracture surface of fish scales after 7 days of demineralization. (b) Higher magnification of the collagen fibers in the boxed area in a. (c) Detailed view of the collagen fibers in the boxed area in b.

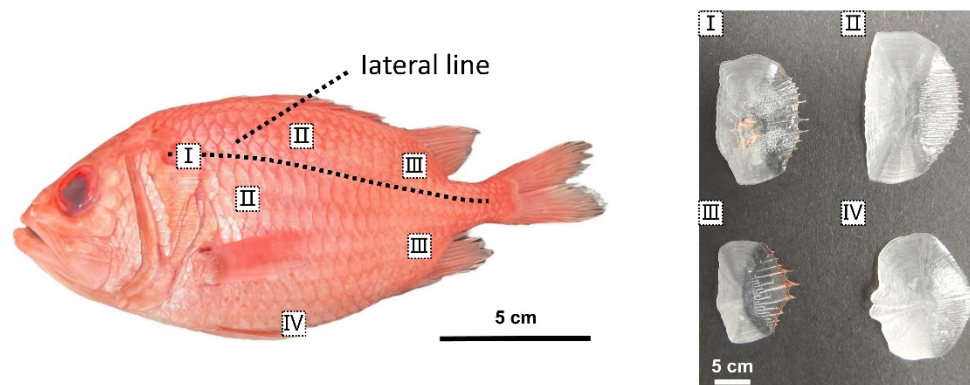


Fig S2. Optical images of scale morphology from different regions of the fish body.

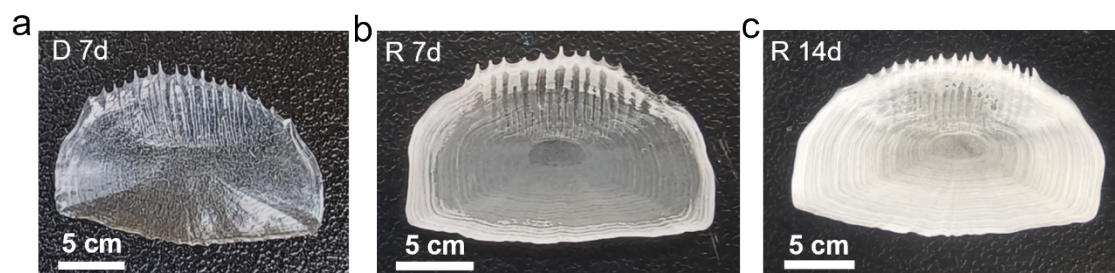
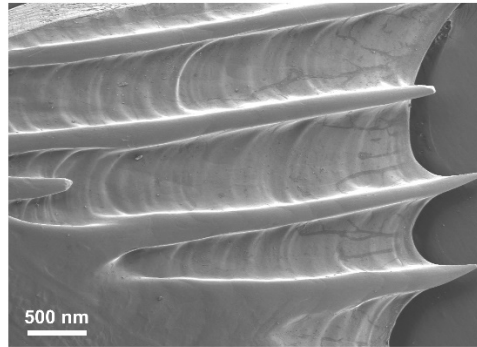
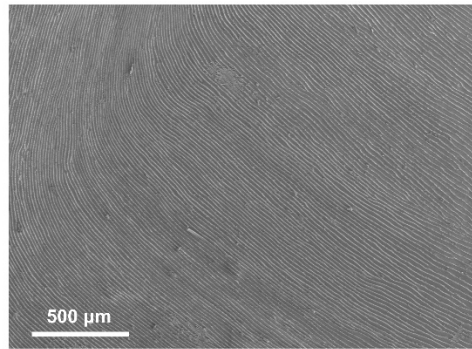


Fig S3. Optical images of fish scales: (a) demineralized for 7 days; (b) after in vitro

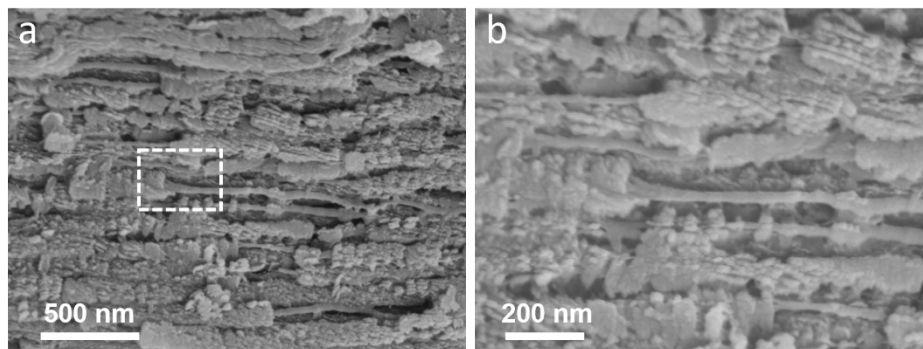
remineralization for 7 days; (c) after in vitro remineralization for 14 days.



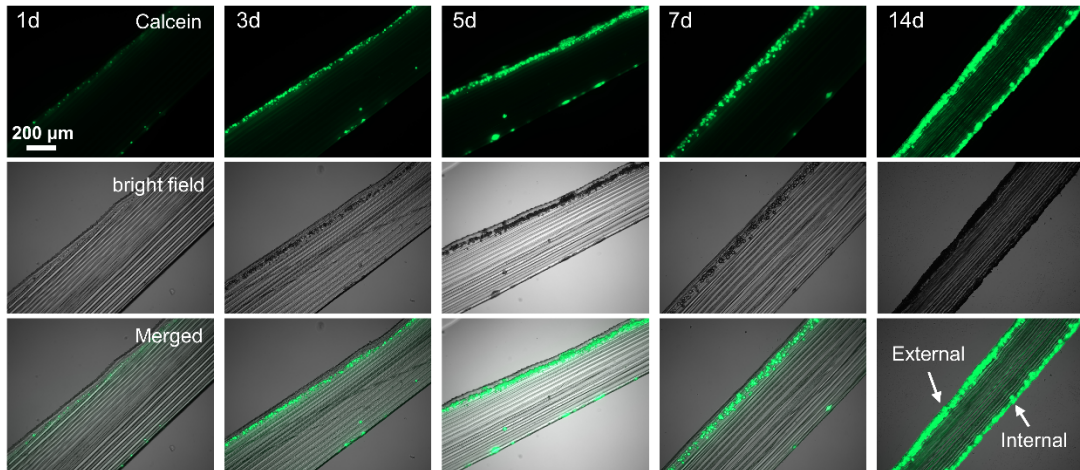
**Fig S4.** SEM image of serrated grooves on the edge of fish scales.



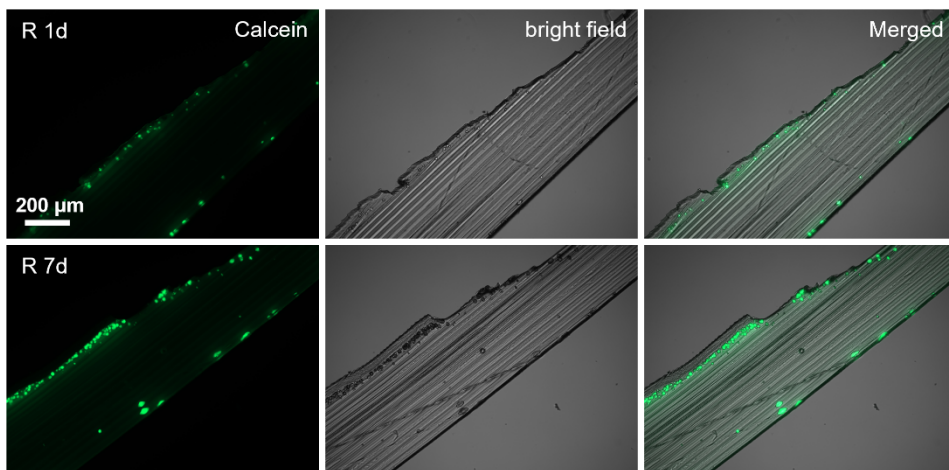
**Fig S5.** SEM image of annular ridges on the external surface of fish scale.



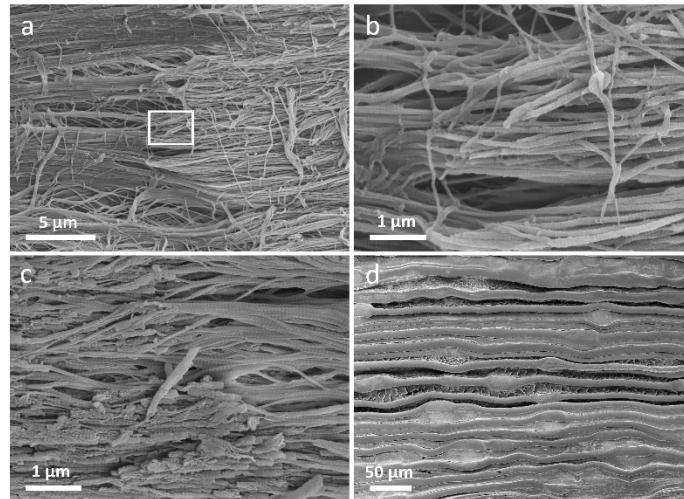
**Fig. S6.** (a) Mineral and collagen fibril distribution in fish scales after 7 days of remineralization. (b) Higher-magnification view of the boxed region in a.



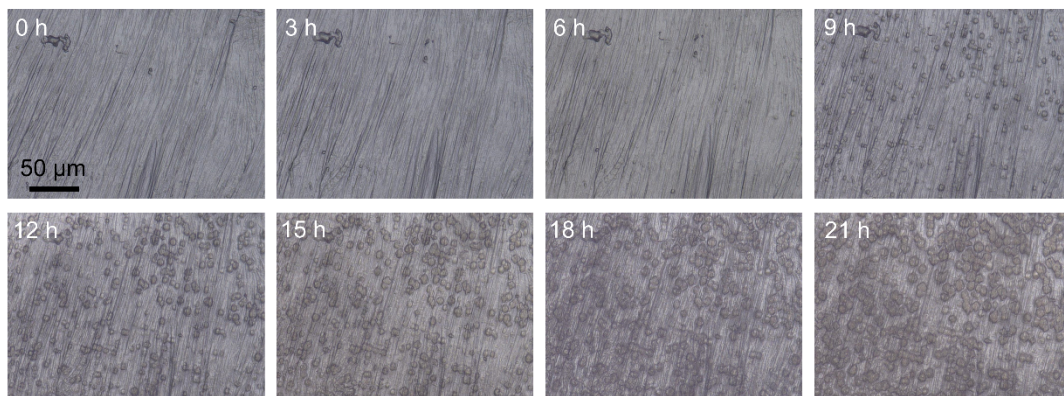
**Fig S7.** Fluorescence images of thin sections of fish scale after mineralization for 1, 3, 5, 7, and 14 days.



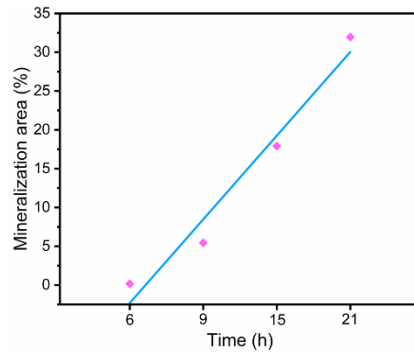
**Fig. S8.** Fluorescence intensity images of fish scale sections after 1 and 3 days of remineralization.



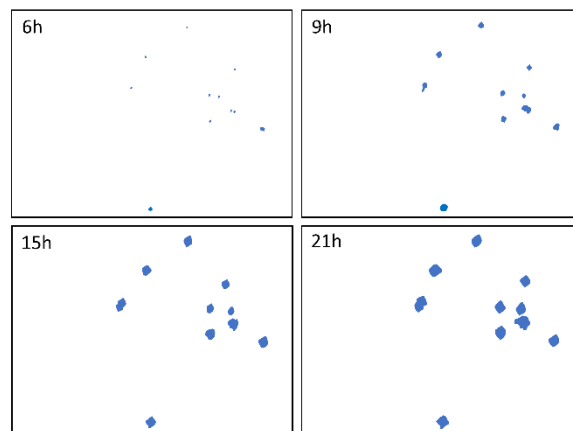
**Fig S9.** Comparative SEM images of mineralized and non-mineralized regions: (a) edge of a mineralized spot; (b) higher-magnification view of the region outlined by the white box in b.; (c) region with incompletely mineralized collagen fibrils; (d) cross-section of a remineralized fish scale.



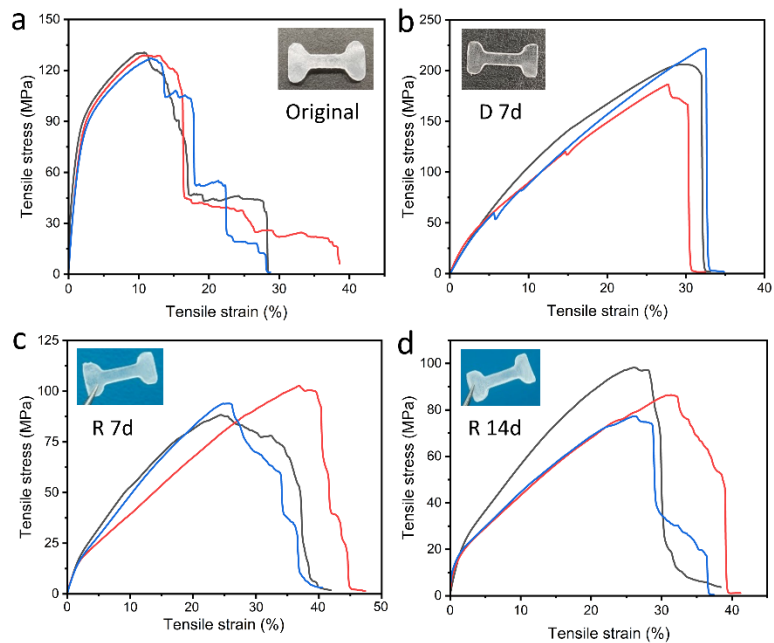
**Fig S10.** Optical images of surface mineralized spots on fish scales after 3-21 h of remineralization.



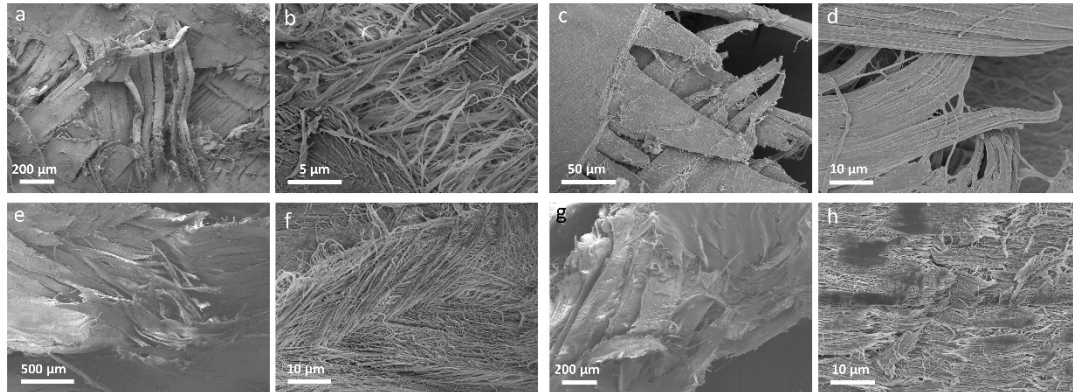
**Fig S11.** Area proportion of mineralized spots during the remineralization process.



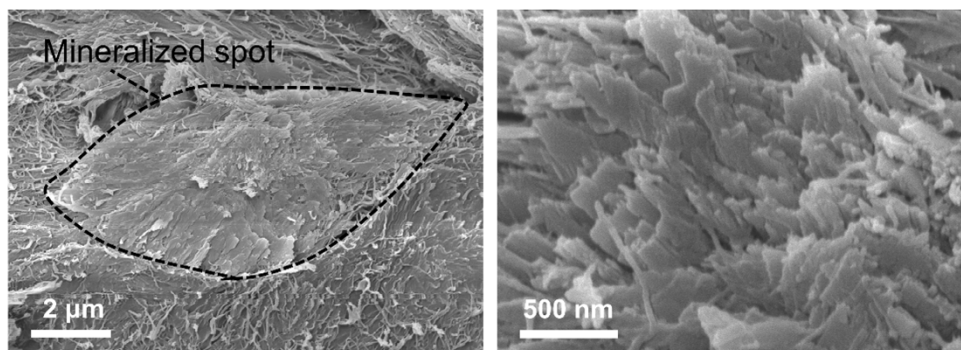
**Fig S12.** Growth of ten individual mineralization spots during 6-21 h of remineralization.



**Fig S13.** Tensile stress-strain curves of (a) original, (b) 7-day demineralized, (c) 7-day remineralized, and (d) 14-day remineralized fish scales.



**Fig S14.** SEM images of tensile fracture morphology of fish scales: (a, b) original; (c, d) demineralized for 7 days; (e, f) remineralized for 7 days; (g, h) remineralized for 14 days.



**Fig S15.** SEM images of mineralized spots and internal mineral crystal morphology

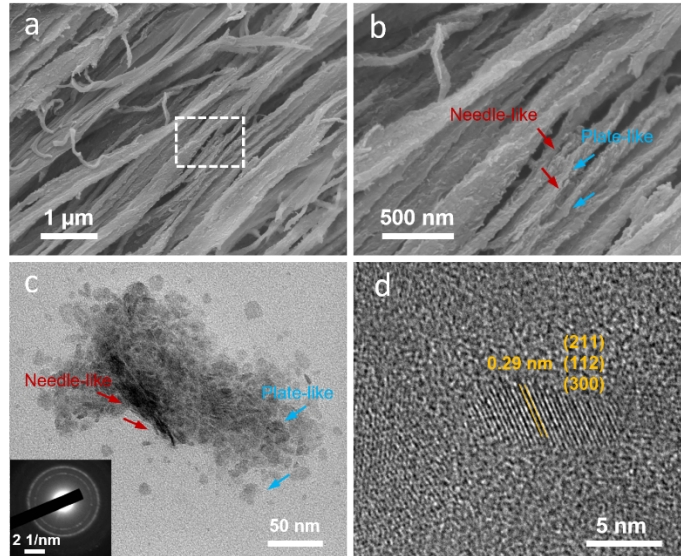
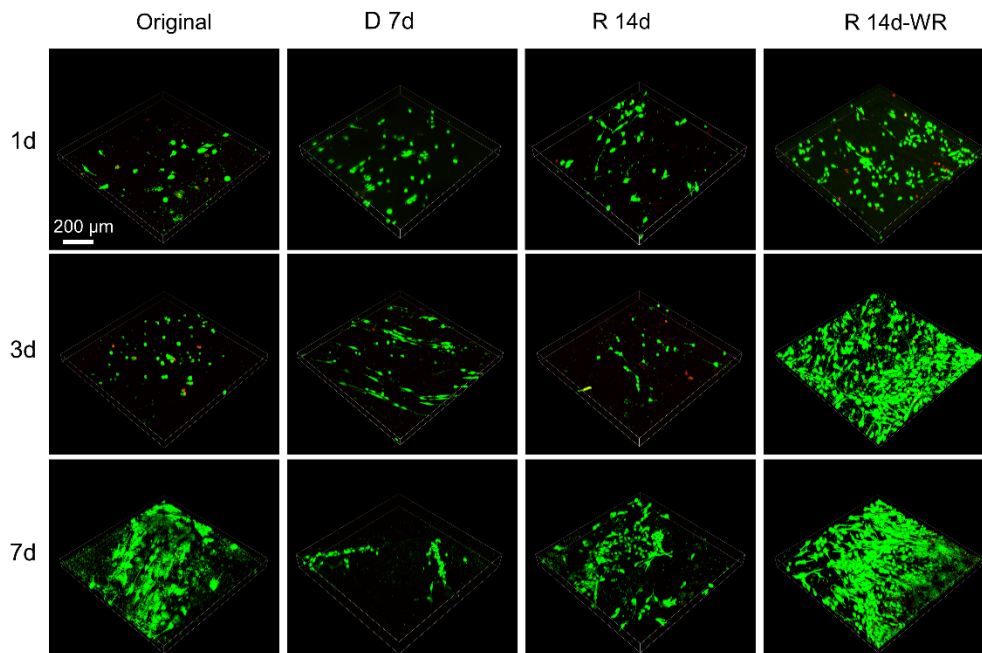
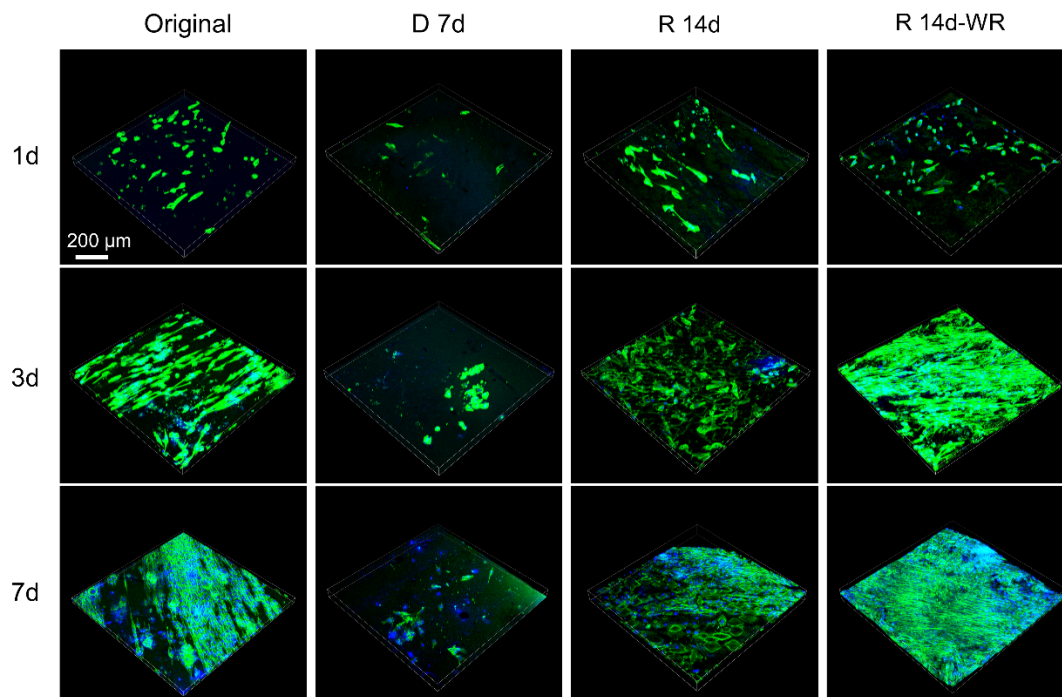


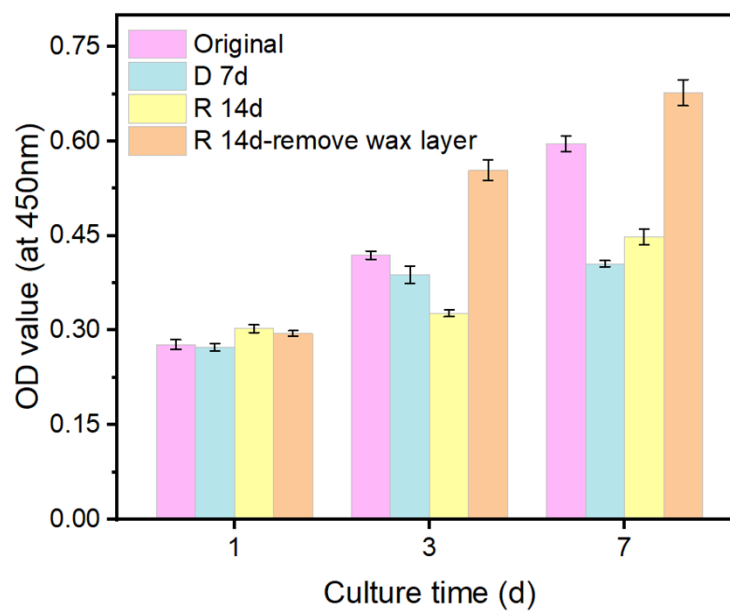
Fig. S16. (a) SEM image showing the distribution morphology of hydroxyapatite within collagen fibrils in the original fish scales. (b) Higher magnification of the area marked by the white frame in a. (c) TEM image showing the distribution morphology of hydroxyapatite within collagen fibrils in the original fish scales. (d) High-resolution image of a single hydroxyapatite nanoplate.



**Fig S17.** Live/dead staining (live cells: green; dead cells: red) of fish scales: original, 7-day demineralized, 7-day remineralized, and 14-day remineralized, after 1, 3, and 7 days.



**Fig S18.** Fluorescence micrographs of cell morphology (cytoskeleton: phalloidin/green; nuclei: DAPI/blue) on fish scales: original, 7-day demineralized, 7-day remineralized, and 14-day remineralized, examined after 1, 3, and 7 days.



**Fig S19.** OD value of CCK-8 of the osteoblasts cultured on the surface of fish scales.

Movie S1. Impact process of a steel ball on a fish scale remineralized for 7 days.

Movie S2. Impact process of a steel ball on a fish scale remineralized for 14 days.

Movie S3. Impact process of a steel ball on an original fish scale.