

Supplementary Information

Deep and Compact Dentinal Tubule Occlusion via Biomimic Mineralization and Mineral Overgrowth

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Supplementary Figures

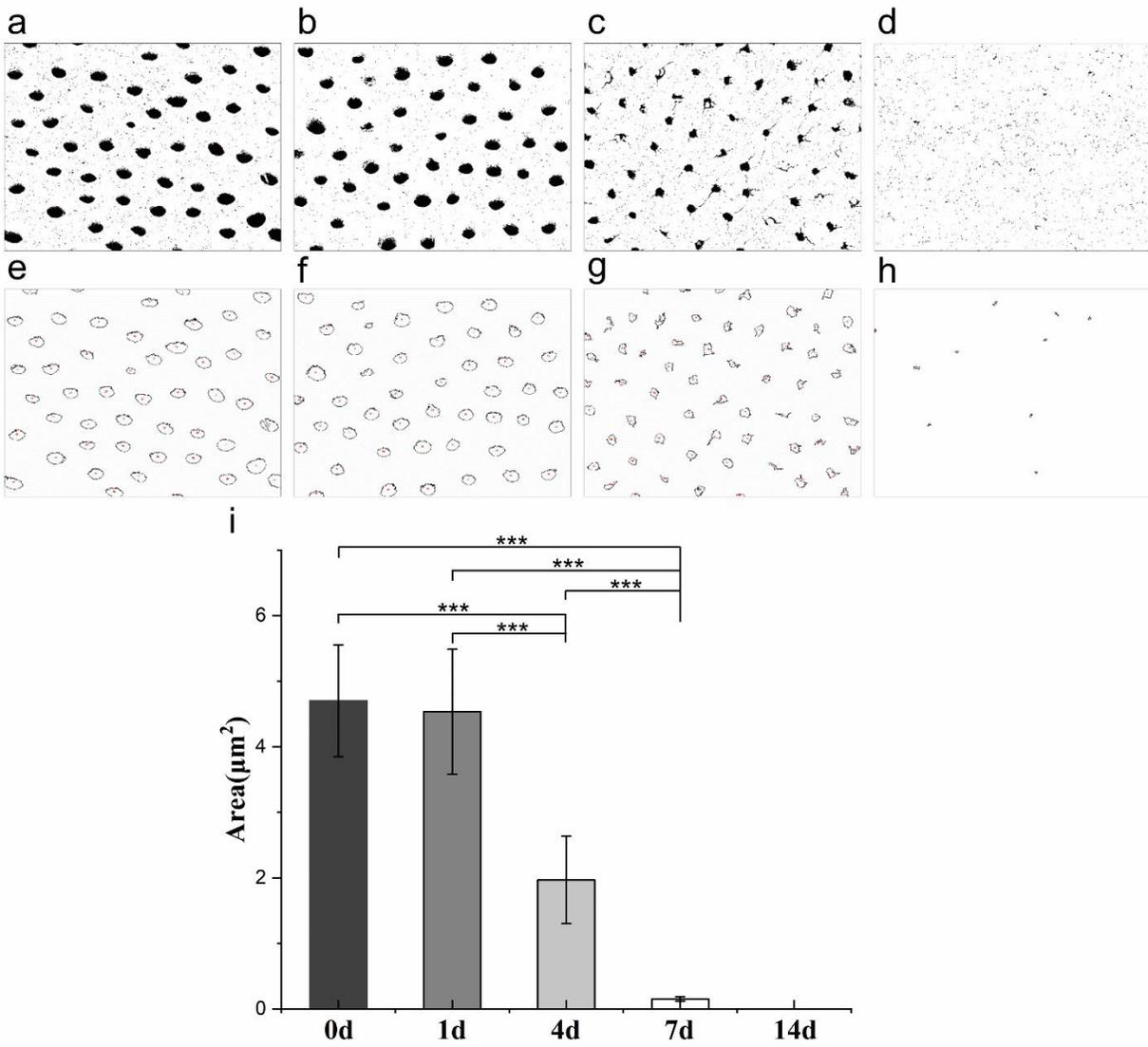


Fig. S1 (a-h) The exposed area of DTs on the top surface of specimens treated with PILP for 0 days (a, e), 1 day (b, f), 4 days (c, g), and 7 days (d, h). (i) Statistical analysis results for the exposed area of tubules. Specimens treated with PILP for 14 days showed no exposed area. *** $P < 0.001$.

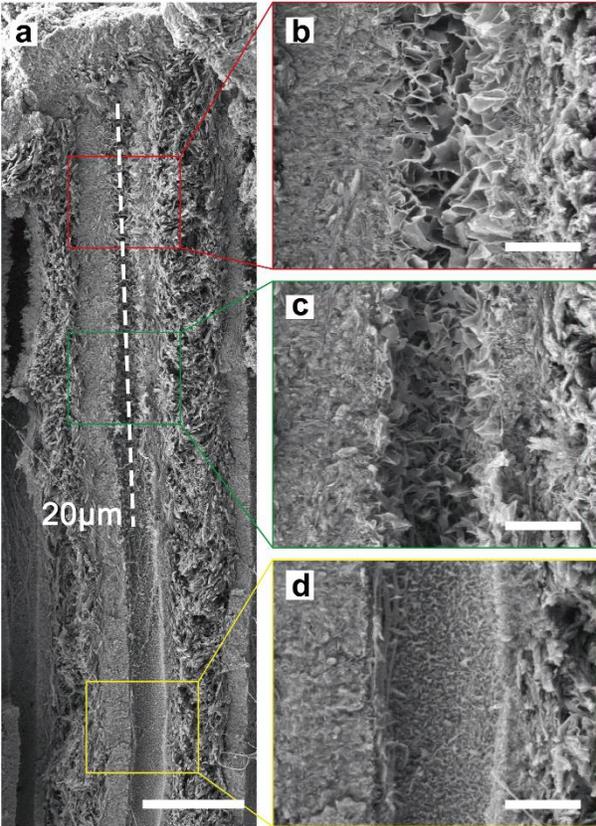


Fig. S2 SEM images showing the mineralized deposition of crystals in the DTs. (a) The crystals occluded the tubule orifice and the mineralized depth reached $19.37 \pm 4.04 \mu\text{m}$ into the DTs. (b) In the superficial layer of the DTs, mineralized sheet-like crystals were completely and densely deposited inside. (c) In the middle layer, mineralized sheet-like crystals deposited around the inner wall of the DT. (d) No mineralized crystals were observed at the base of DTs. Scale bars: 5 μm (a), 1 μm (b-d),

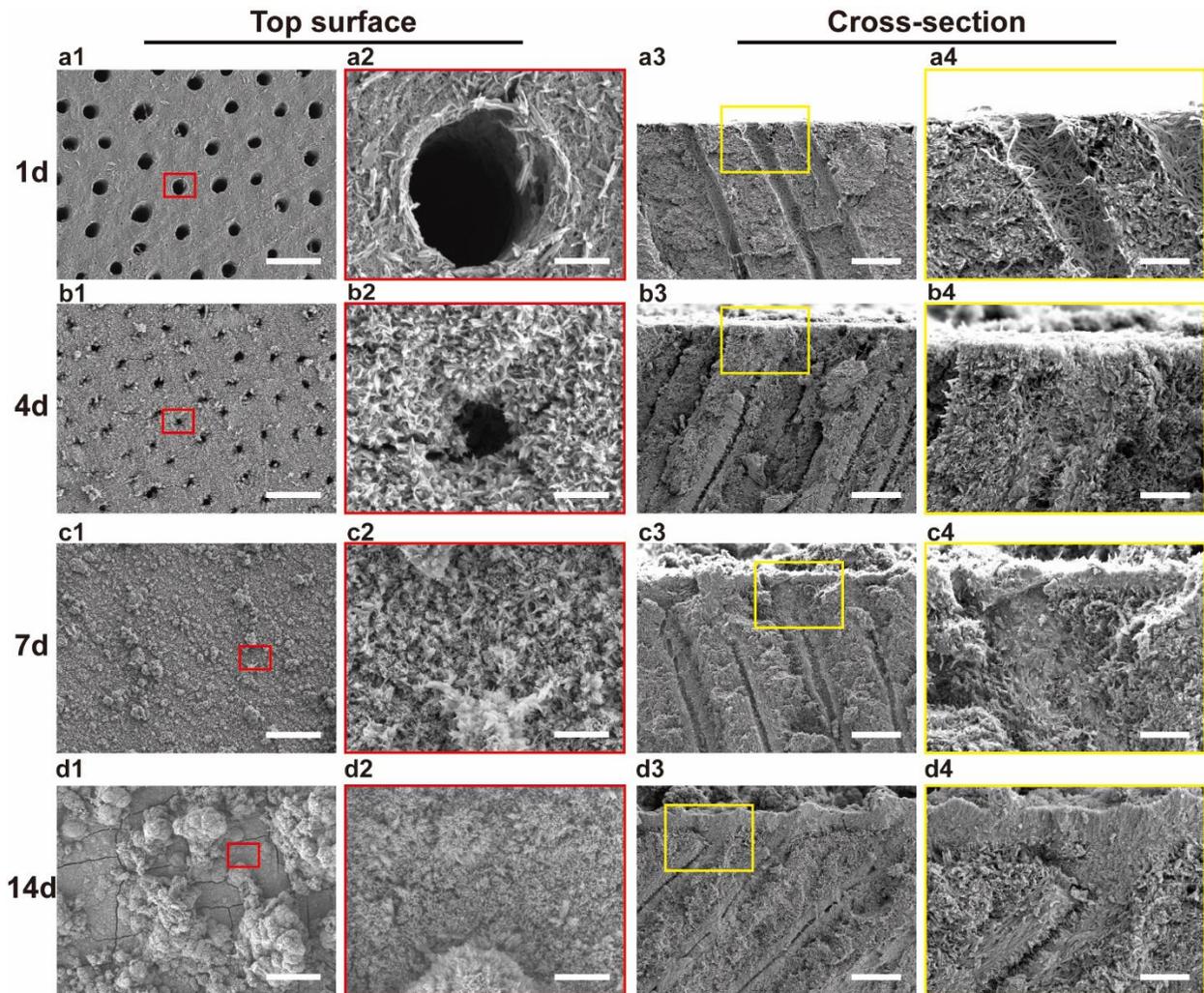


Fig. S3 SEM micrographs of the top surfaces and cross-sections of dentin discs immersed in PILP (m-SBF and p-Asp of $240 \mu\text{g mL}^{-1}$) for 1 day (a1-a4), 4 days (b1-b4), 7 days (c1-c4), 14 days (d1-d4) before ultrasonication. (a1-a4) Specimens treated with PILP for 1 day showed the surface of dentin discs was relatively clean with only trace amounts of needle-shaped deposition. (b1-b4) Specimens treated with PILP for 4 days showed crystal deposits in the superficial layer of the DTs, and the DT orifices were significantly narrowed. (c1-c4) Specimens treated with PILP for 7 days showed complete closure of the DT orifices with deposition deep into the DT. (e1-e4) Specimens treated with PILP for 14 days showed that a large number of crystals were compactly and densely formed on the surface of dentin disc and in the deep layer of DTs,

completely occluding the tubule orifices. Scale bars: 10 μm (a1-d1), 1 μm (a2-d2), 6 μm (a3-d3),
2 μm (a4-d4).

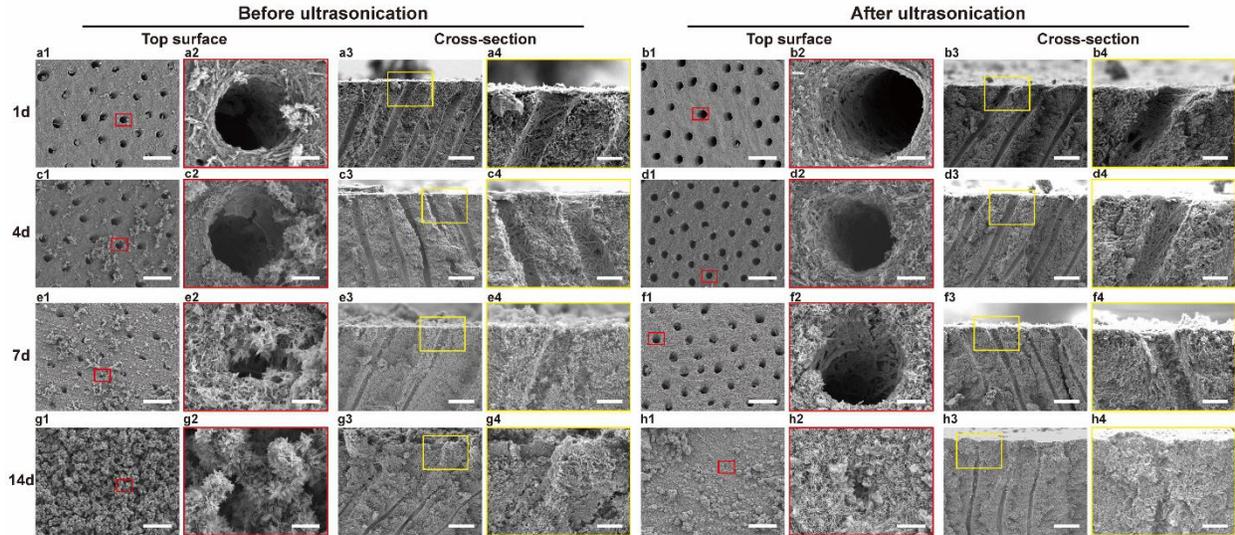


Fig. S4 SEM micrographs of the top surfaces and cross-sections of dentin discs immersed in PILP (m-SBF and p-Asp of $50 \mu\text{g mL}^{-1}$) for 1 day (a1-b4), 4 days (c1-d4), 7 days (e1-f4), 14 days (g1-h4) before or after ultrasonication. Specimens treated with PILP for 1 day (a1-b4) showed complete opening of the DTs. Specimens treated with PILP for 4 days (c1-c4) and 7 days (e1-e4) showed a thin layer of minerals on the surface of dentin discs and the inner wall of narrowed DTs, which basically disappeared after ultrasonication (d1-d4, f1-f4). Specimens treated with PILP for 14 days (g1-g4) showed complete occlusion of the tubule orifice with deep crystal deposits in the DTs, as well as partial crystal deposited on the surface and inside the tubules after ultrasonication (h1-h4). Scale bars: $10 \mu\text{m}$ (a1-h1), $1 \mu\text{m}$ (a2-h2), $6 \mu\text{m}$ (a3-h3), $2 \mu\text{m}$ (a4-h4).

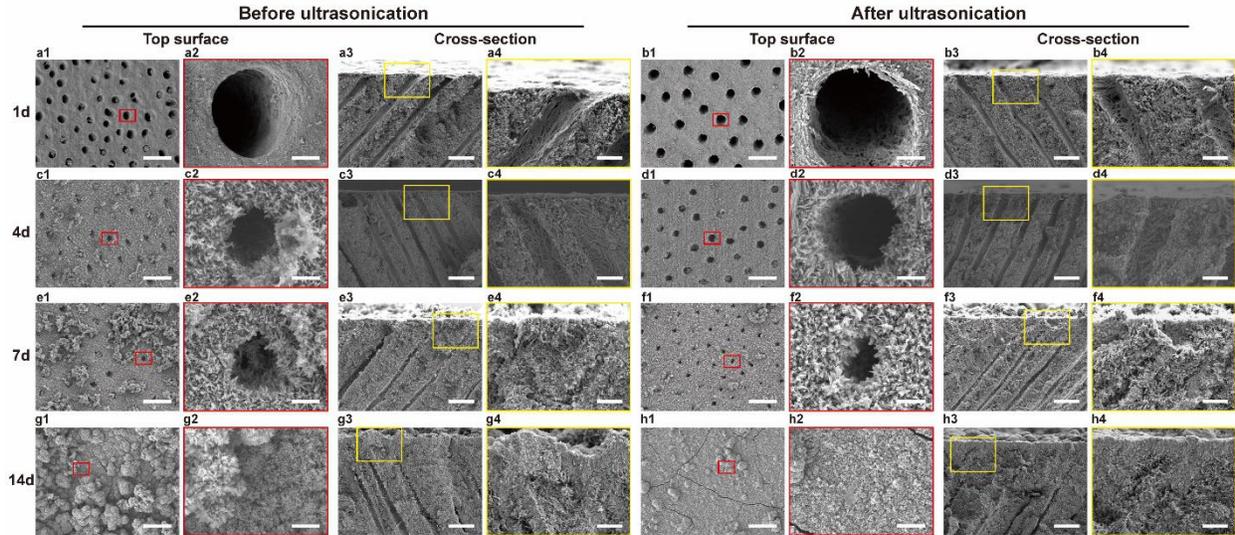


Fig. S5 SEM micrographs of the top surfaces and cross-sections of dentin discs immersed in PILP (m-SBF and p-Asp of $120 \mu\text{g mL}^{-1}$) for 1 day (a1-b4), 4 days (c1-d4), 7 days (e1-f4), 14 days (g1-h4) before or after ultrasonication. Specimens treated with PILP for 1 day (a1-b4) showed no clear deposits on the surface or within the DTs before and after ultrasonication, and the DT diameter was unchanged. Specimens treated with PILP for 4 days (c1-c4) showed the mineral sparsely deposited on the surface of dentin discs, which were largely removed after ultrasonication (d1-d4). Specimens treated with PILP for 7 days (e1-e4) showed uneven needle-like deposits on the surface of the dentine discs, which closed some of the tubule orifices, and some deposits were sloughed off after ultrasonication and the DTs opened narrowly (f1-f4). Specimens treated with PILP for 14 days (g1-g4) showed a dense layer of crystal deposits on the surface of the dentin discs that penetrated deep into the tubule interior, and the occlusion of dentin tubules didn't disappear after ultrasonication (h1-h4). Scale bars: $10 \mu\text{m}$ (a1-h1), $1 \mu\text{m}$ (a2-h2), $6 \mu\text{m}$ (a3-h3), $2 \mu\text{m}$ (a4-h4).

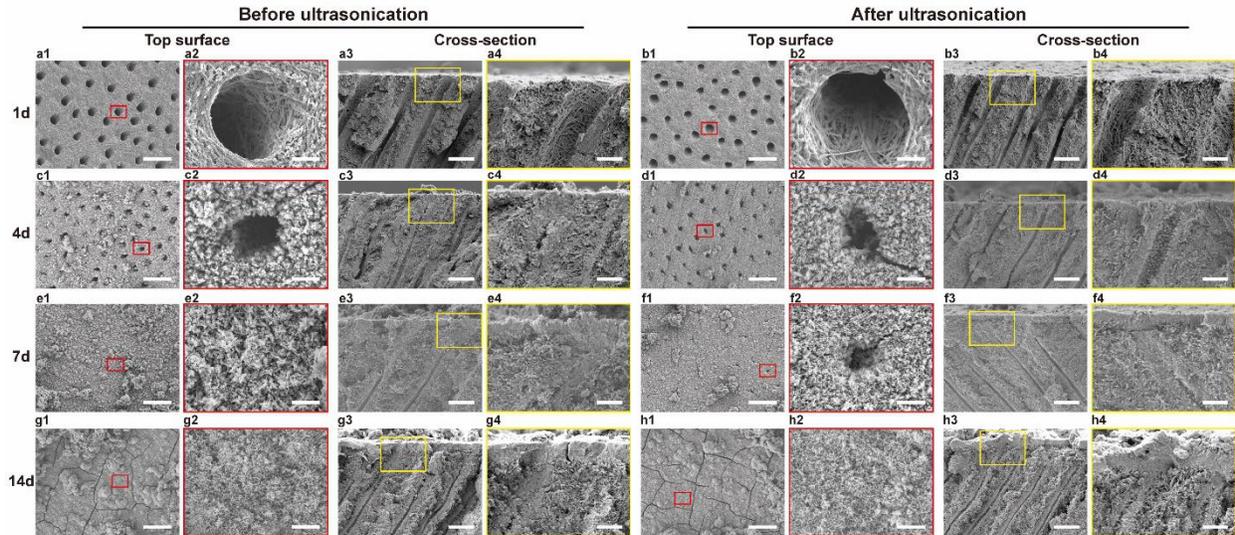


Fig. S6 SEM images showing the top surfaces and cross-sections of dentin discs immersed in PILP (m-SBF and p-Asp of $600 \mu\text{g mL}^{-1}$) for 1 day (a1-b4), 4 days (c1-d4), 7 days (e1-f4), 14 days (g1-h4) before or after ultrasonication. Specimens treated with PILP for 1 day (a1-b4) showed that there was no deposit sediment on the surface of dentin disc and the orifice was completely open. Specimens treated with PILP for 4 days (c1-c4) showed sparse mineral deposits on the surface of the dentine discs and around the tubule orifices, which were mostly sloughed off after ultrasonication (d1-d4). Specimens treated with PILP for 7 days (e1-e4) showed a uniform crystal mineral deposition on the surface of the dentine discs, occluding the tubule orifices, and some deposits were removed after ultrasonication so that a small number of tubule orifices were opened (f1-f4). Specimens treated with PILP for 14 days (g1-g4) showed densely distributed massive crystal mineral deposits on the surface of the dentine discs and inside the DTs, and the tubule orifice remained all closed after ultrasonication (h1-h4). Scale bars: $10 \mu\text{m}$ (a1-h1), $1 \mu\text{m}$ (a2-h2), $6 \mu\text{m}$ (a3-h3), $2 \mu\text{m}$ (a4-h4).

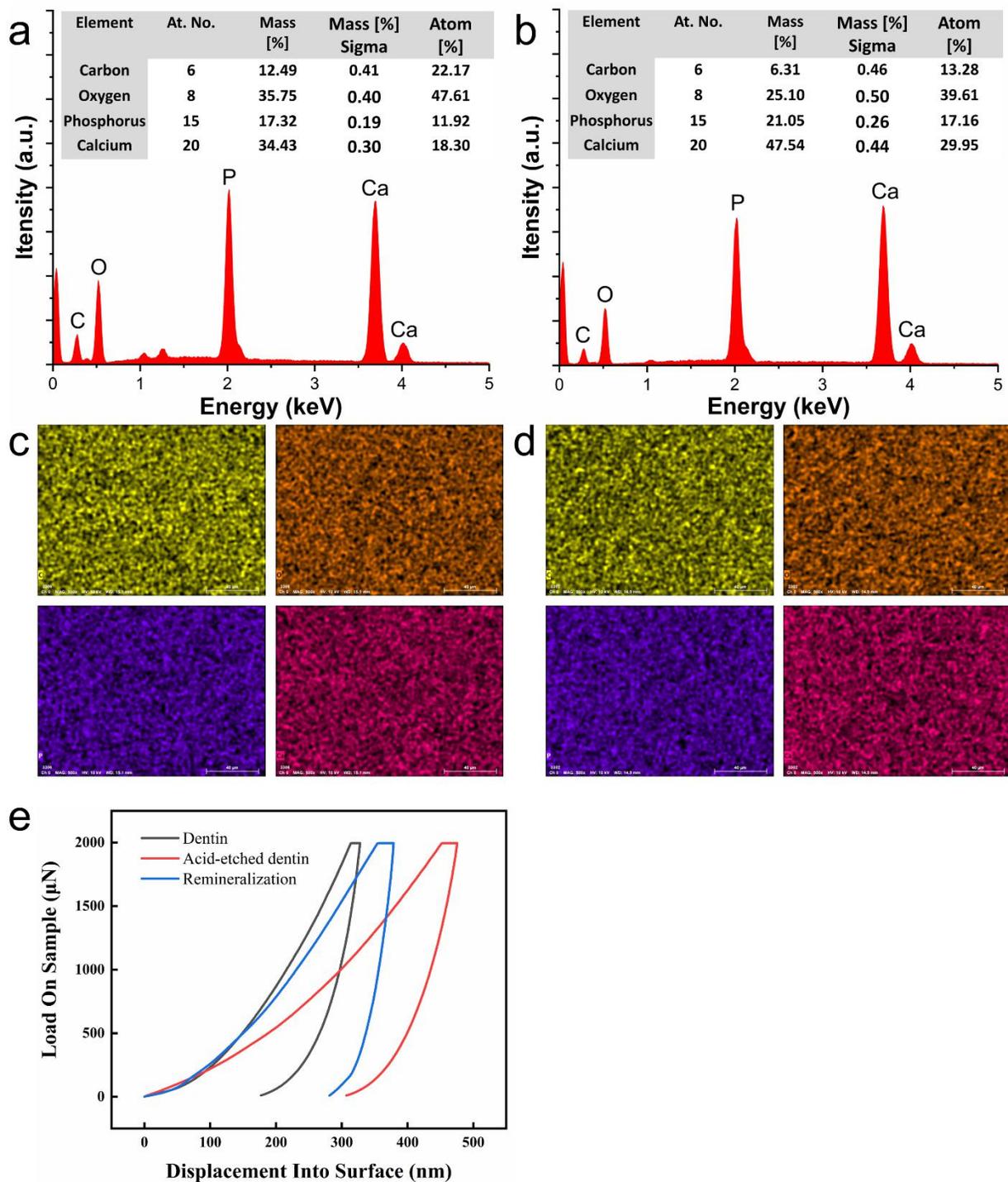


Fig. S7 (a, b, c, d) EDS results for native dentin (a, c) and the remineralization layer on the surface of PILP-treated dentin (b, d). The Ca/P ratio of the remineralization layer was 1.75, which is consistent with that of HAP. C (yellow), O (orange), P (purple), Ca (pink) were uniformly

distributed on the dentin surface. (e) Loading-unloading curves obtained by the nanoindentation tests at a maximum loading force of 2000 μN .

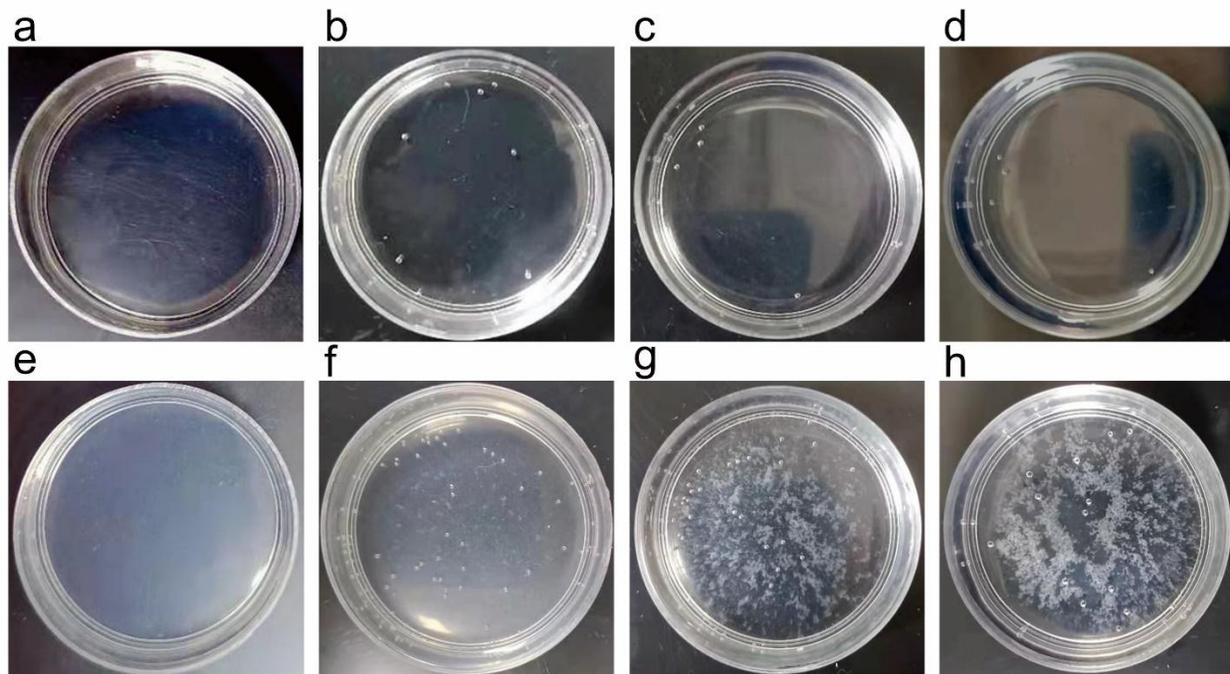


Fig. S8 Appearance of PILP and m-SBF. (a-d) Photographs of PILP at 0 hours (a), 3 hours (b), 12 hours (c), and 24 hours (d). (e-h) Photographs of m-SBF at 0 hours (e), 1 hour (f), 2 hours (g), and 12 hours (h).

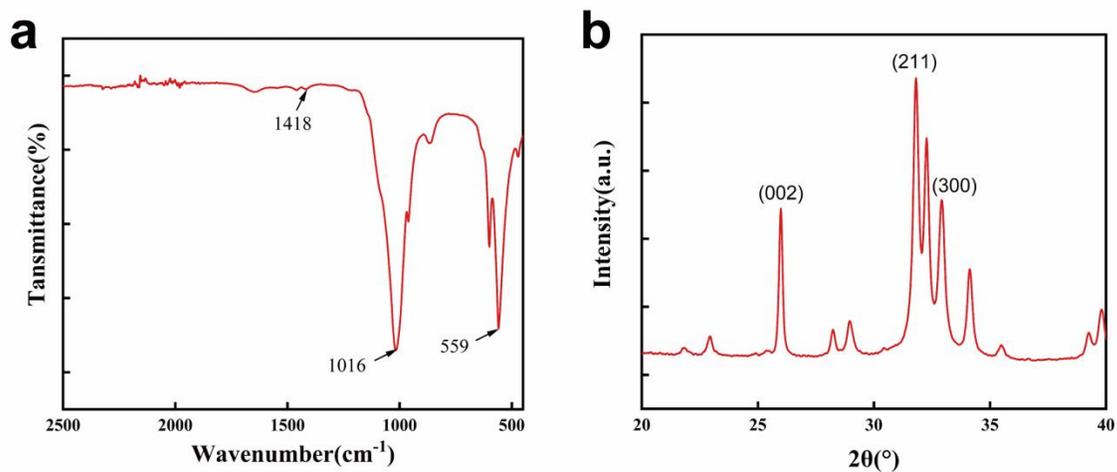


Fig. S9 Characterization of dentin treated with m-SBF for 14 days before ultrasonication. (a) XRD results for m-SBF treated dentin before ultrasonication (14 days). (b) FTIR-ATR results for m-SBF treated dentin before ultrasonication (14 days).

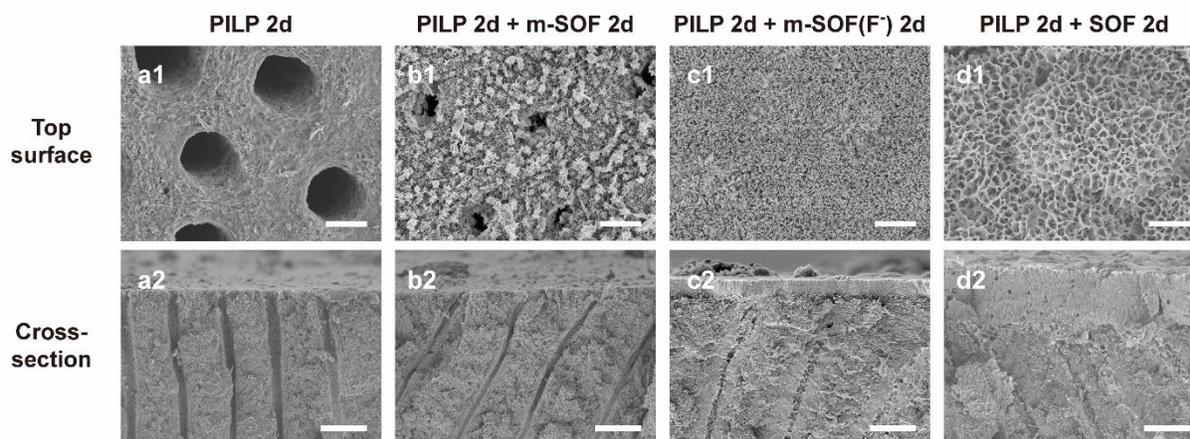


Fig. S10 SEM images depicting accelerated remineralization of dentin in different solutions. The top surfaces and cross-sections of dentin discs immersed in PILP for 2 days(a1-a2) and in m-SOF (b1-b2), m-SOF(F⁻) (c1-c2) and SOF (d1-d2) for another 2days. (a1-a2) Specimens treated with PILP for 2 days showed newly formed crystals did not block the DTs. (b1-b2) Specimens treated with PILP for 2 days and m-SOF for another 2 days showed crystals deposits in the superficial layer of the DTs, narrowing the diameter of DT orifice. (c1-c2) Specimens treated with PILP for 2 days and m-SOF(F⁻) for another 2days showed crystals formed a layer of dense and needle shaped deposition on the surface of dentin disc and almost blocked DTs. (d1-d2) Specimens treated with PILP for 2 days and SOF for another 2days showed crystals uniformly dispersed and densely deposited on the surface of dentin disc and in the deep layer of DTs, completely occluding the DTs. Scale bars: 2 μm (a1-d1), 6 μm (a2-d2).