

Supplementary Materials

Fractal-like hierarchical organization of bone begins at the nanoscale

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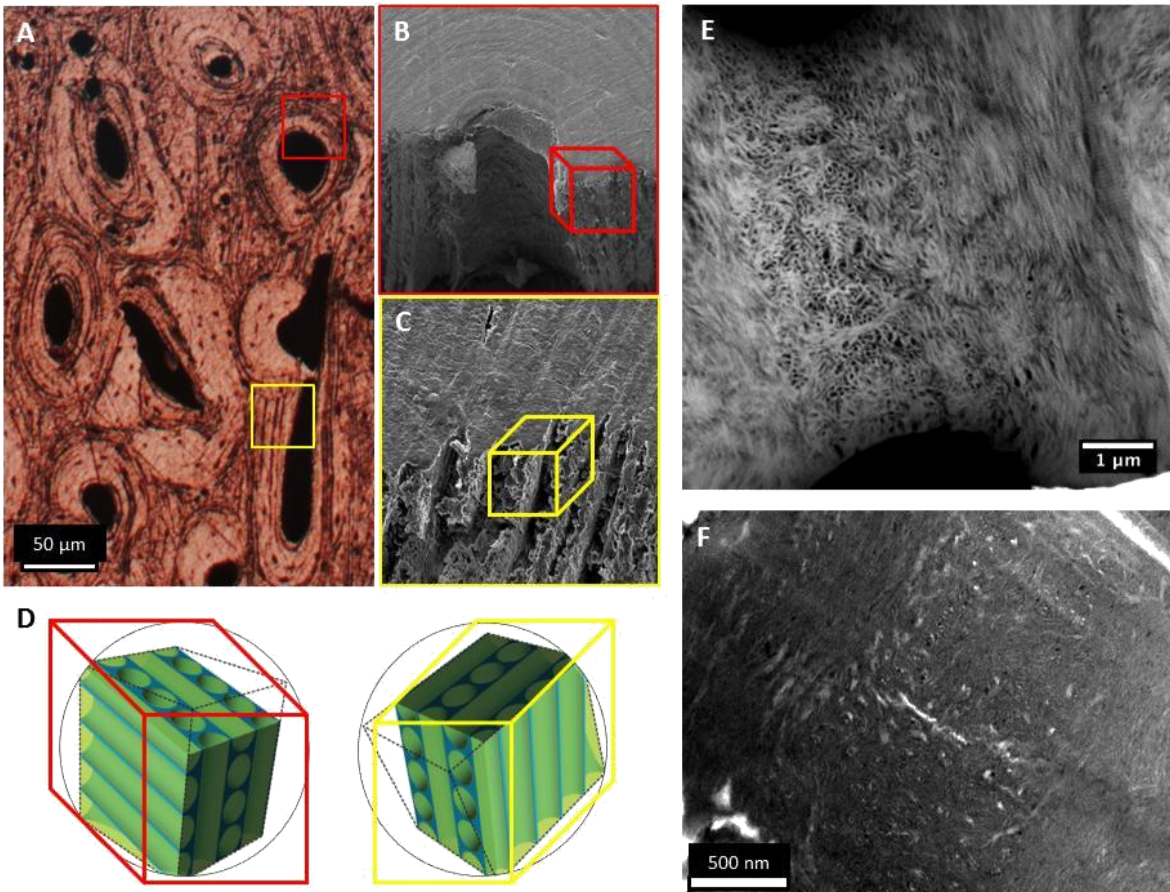


Fig. S1. Geometry of the specimens used in this study. (A) Light microscope image of a polished section of unmodified human compact bone. The majority of osteons are cut in cross-section, except one osteon in the bottom right that is oriented longitudinally. (B) and (C) correspond to the red and yellow squares in the panel (A), respectively. These are SEM images of osteonal bone fractured across and along the osteon axes. Note that both show lamellar structure. The schematic in (D) shows that if a sample is prepared perpendicular to lamellar arrays, either across or along an osteon, the resulting 2D image will display alternation of in-plane (filamentous) and out-of-plane (lacey or spiral) arrays of mineralized collagen fibrils. This can be observed at higher magnification in dark field STEM (E) or bright field TEM (F), for example. The schematic in panel D is adapted from Reznikov et al., *Bone* (2014) (28).

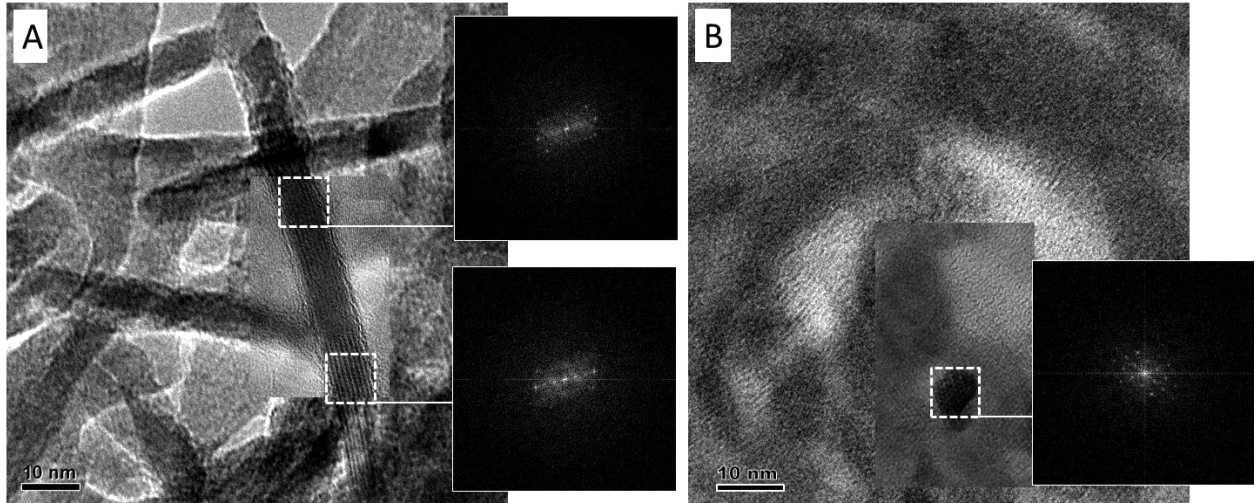


Fig. S2 (A) HRTEM micrograph of a few bone crystals taken from the lacey motif. The dashed boxes along the oblique crystal were processed using fast Fourier transform, which demonstrates similar patterns slightly tilted around the zone axis. Note, that no {002} lattice fringes could be identified in agreement with the SAD pattern of this motif (see Fig. 1A'). (B) HRTEM micrograph of an edge-on crystal from the rosette motif showing 2D lattice image of apatite with c-axis parallel to the zone axis: fast Fourier transform of the boxed area reveals the hexagonal crystal symmetry and the absence of {002} lattice fringes in accordance with the SAD patterns recorded from this motif (see Fig. 1C').

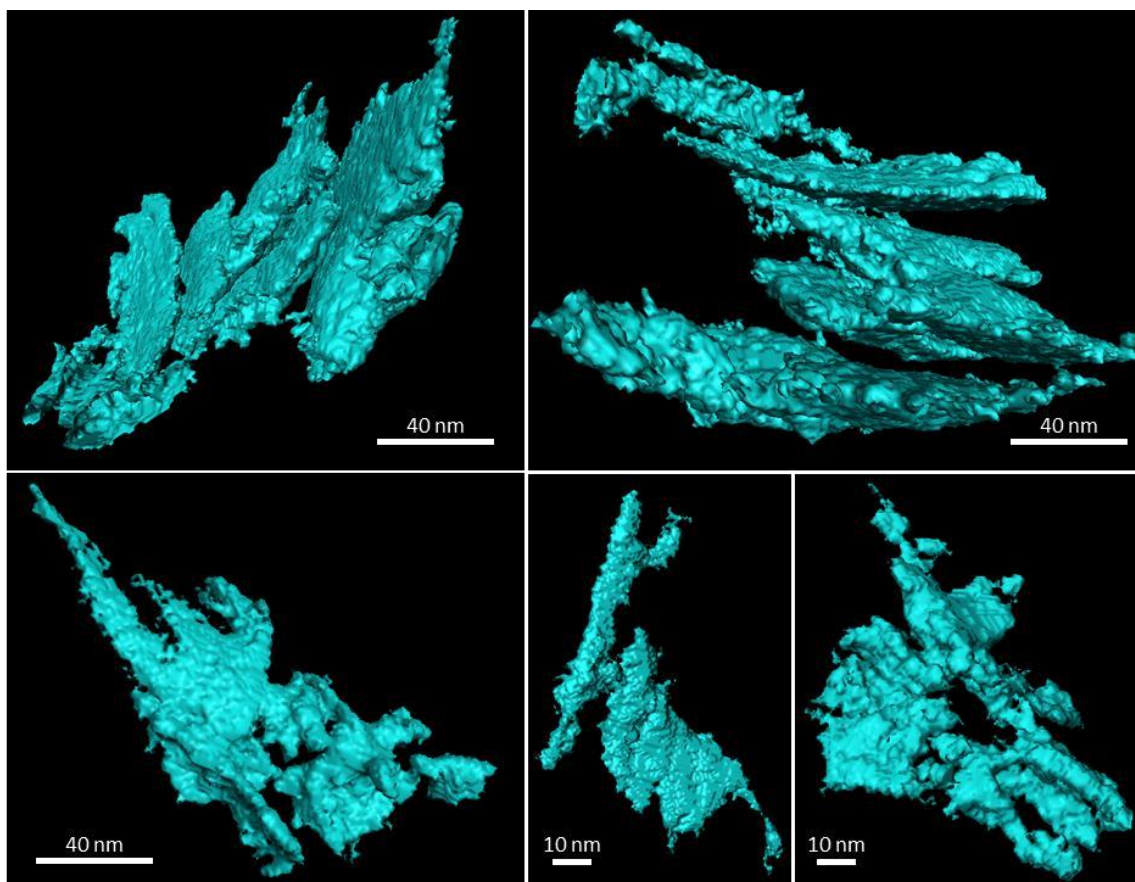


Fig. S3 3D rendering of individual mineral entities in the tomogram from Fig. 3 (lacey pattern only). Note hierarchical assembly of acicular crystallites into irregular slightly twisted platelets, stacks of parallel platelets and wedged aggregates of platelet stacks.

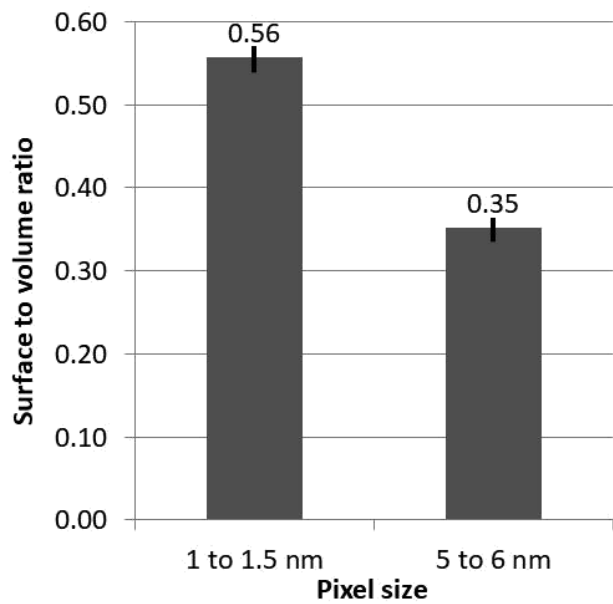


Fig. S4 Comparison of the surface to volume ration between the original tomogram resolution and an artificially coarsened volume with a pixel size 5-6 nm.