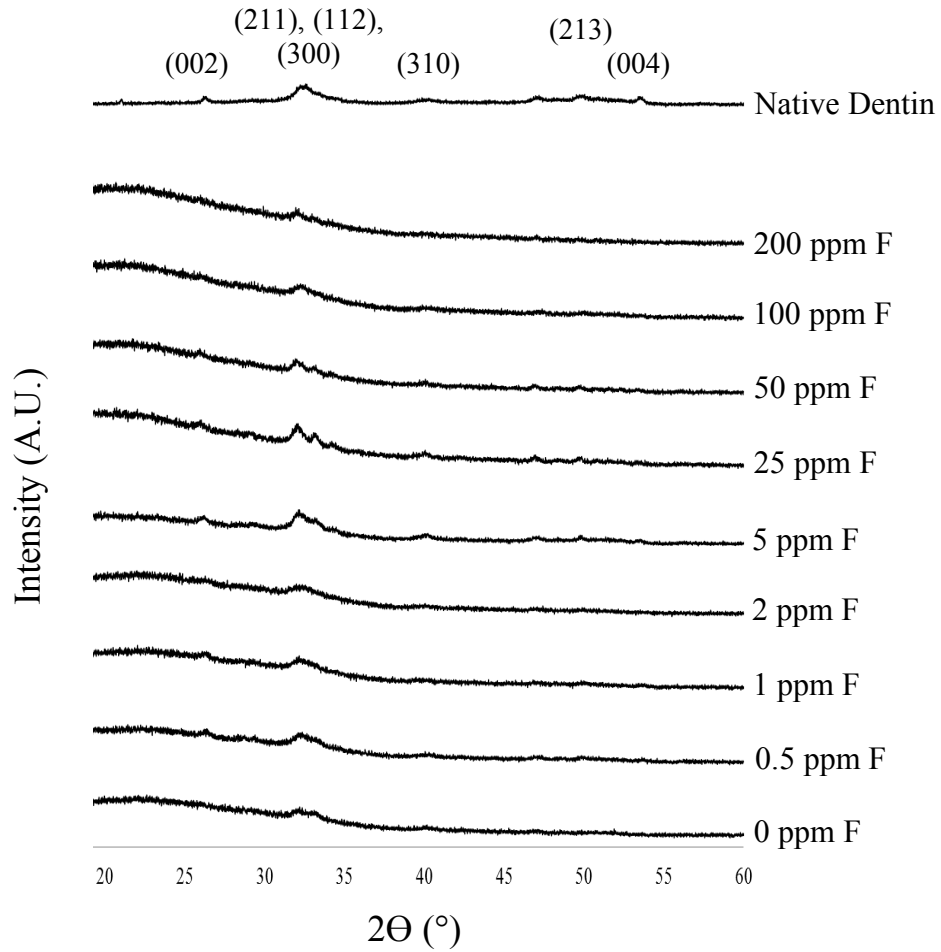
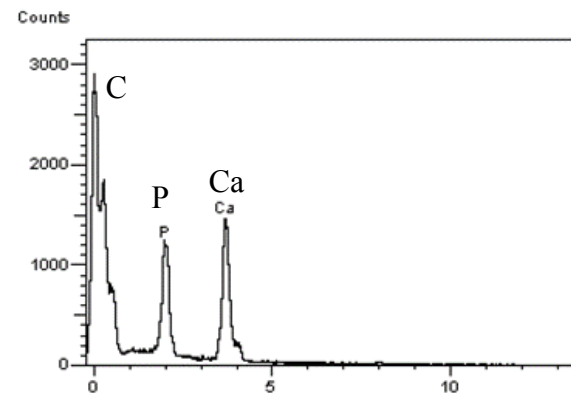
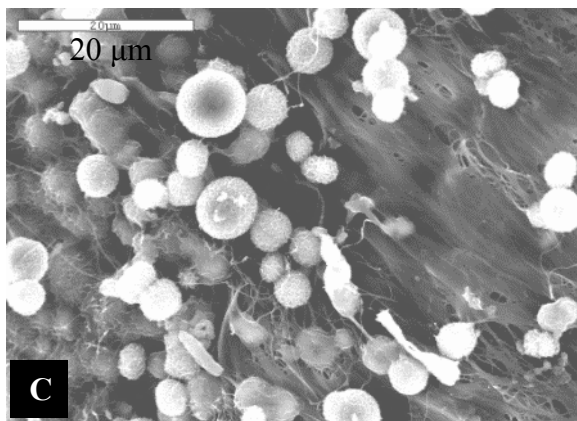
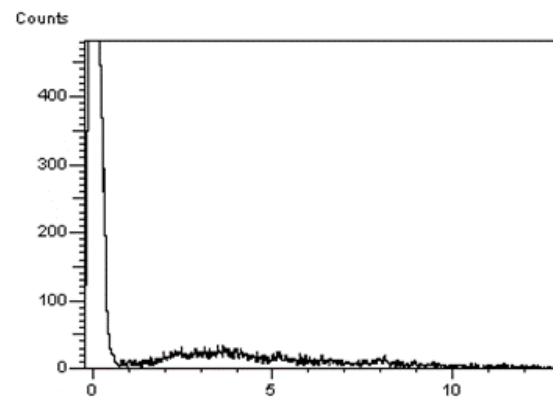
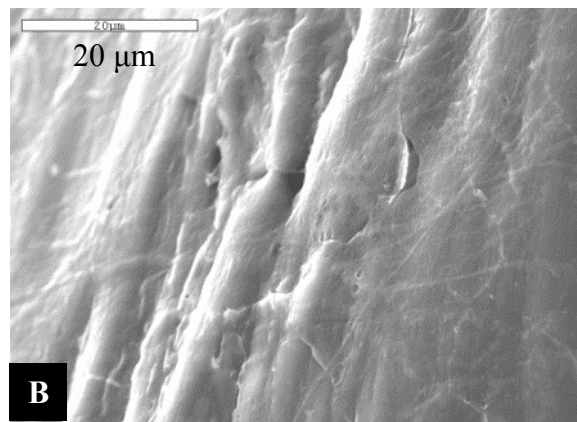
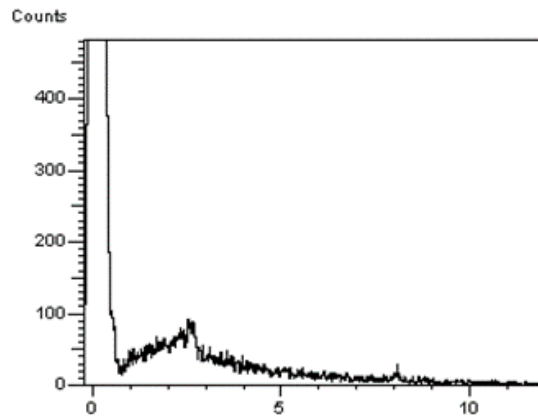
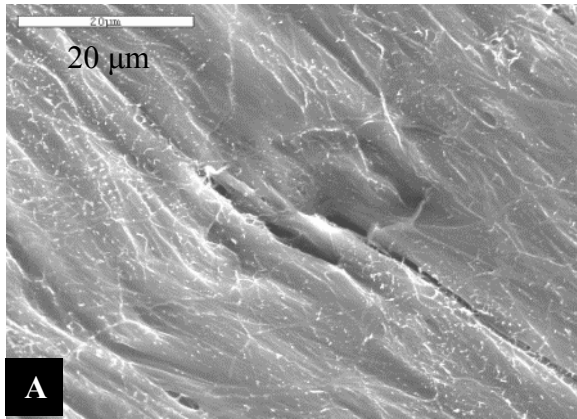


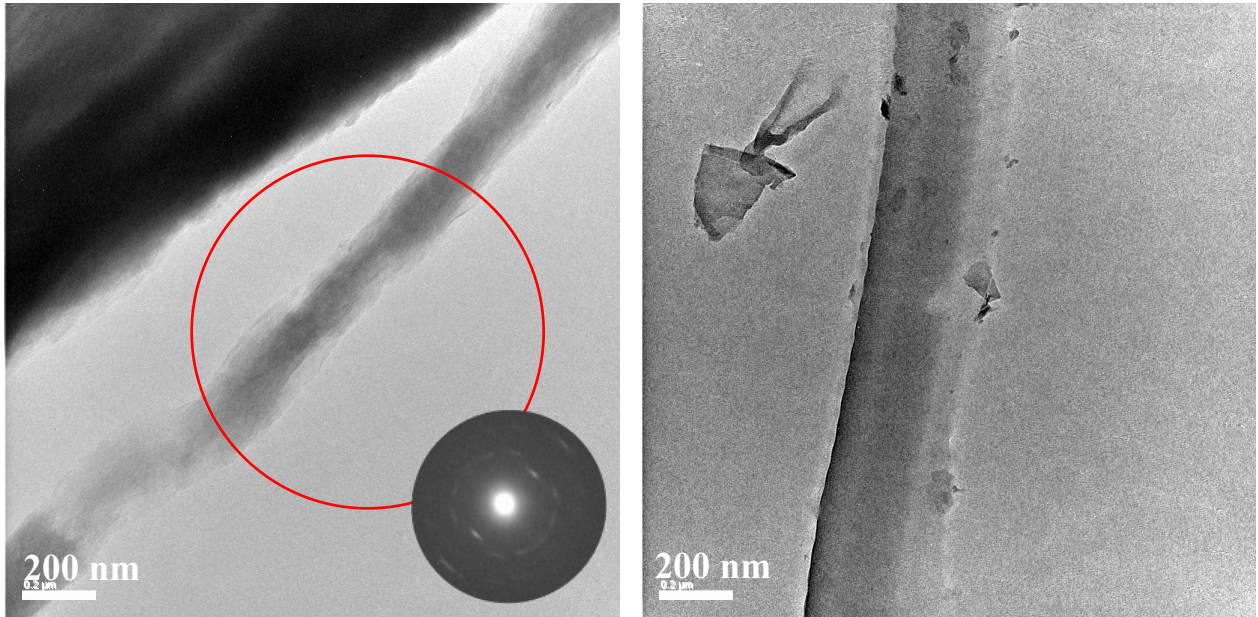
## Supplemental Information



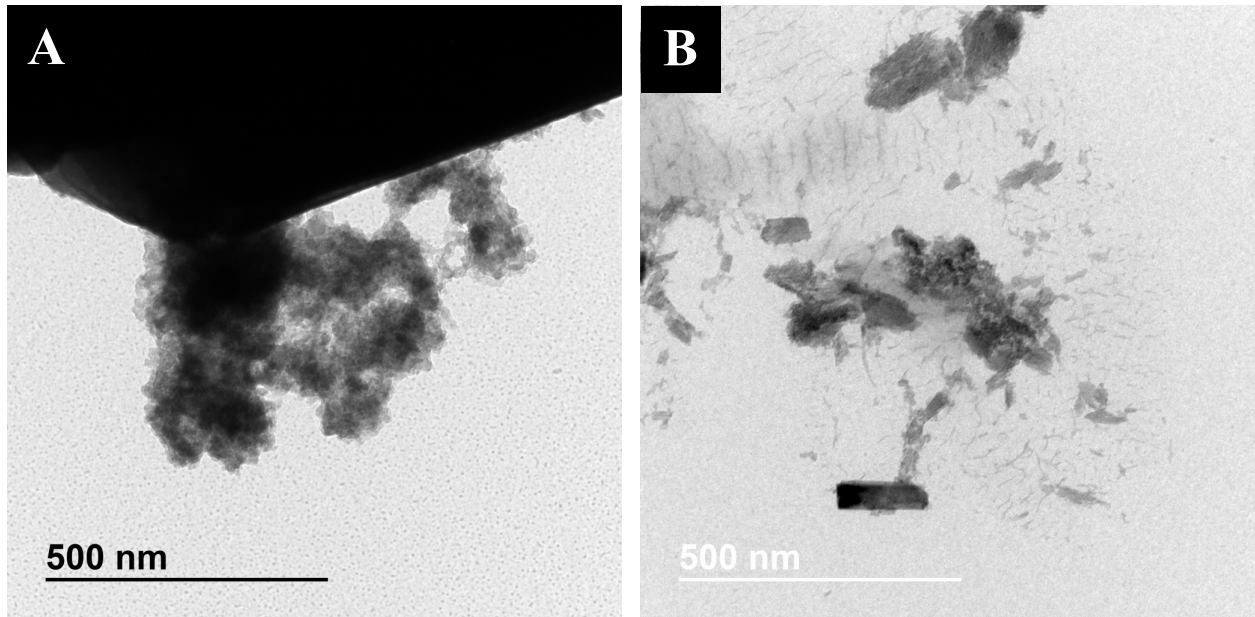
**Supplemental Figure 1. X-ray diffraction patterns of native dentin and mineralized samples. All patterns confirm the presence of nanocrystalline/impure, apatitic mineral.**



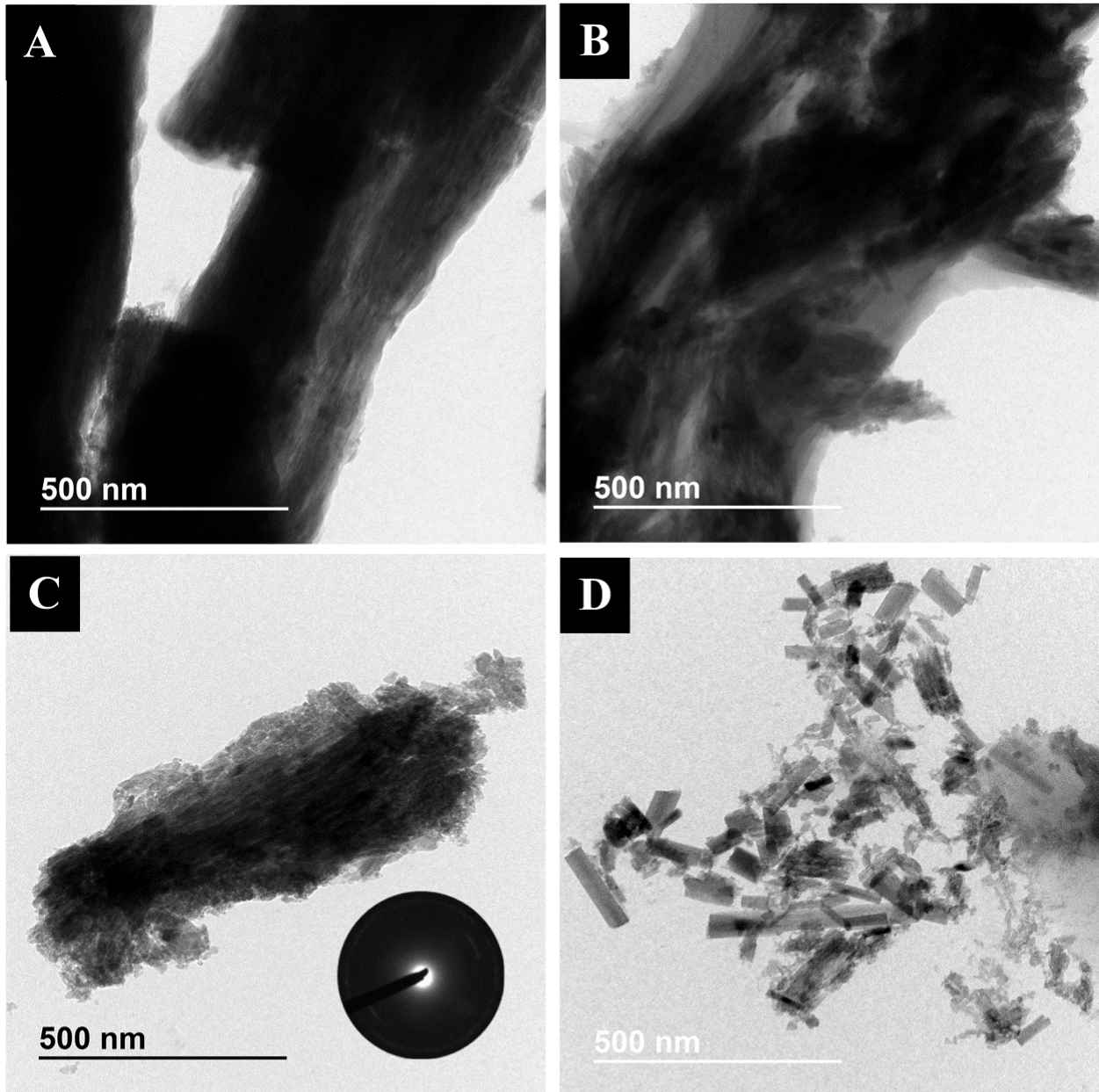
**Supplemental Figure 2. SEM micrographs (left) and their corresponding EDS spectra (right) of unmineralized rat tail tendon before EDTA treatment (A), after EDTA treatment (B), and rat tail tendon mineralized by the conventional process (without the addition of pAsp or fluoride) (C). The fibrils in unmineralized tendon dehydrate into a smooth mat after freeze drying. Mineralization without a process-directing agent such as pAsp leads to hydroxyapatite spherulites on the surfaces of the tendon and little or no intrafibrillar mineral.**



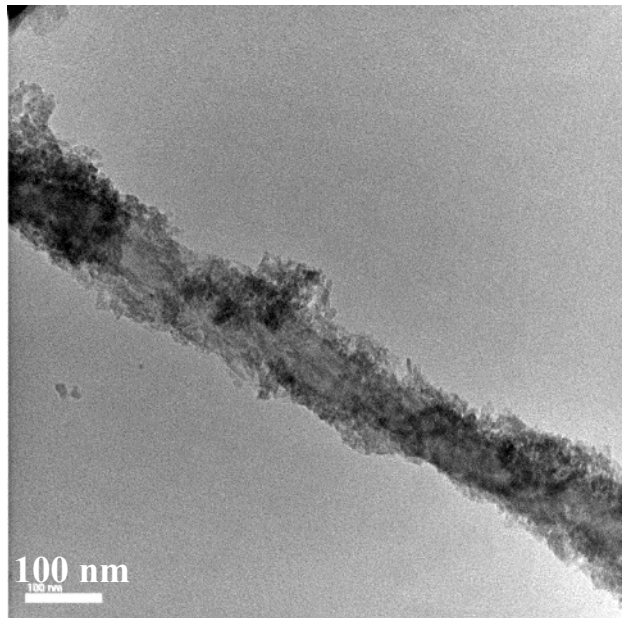
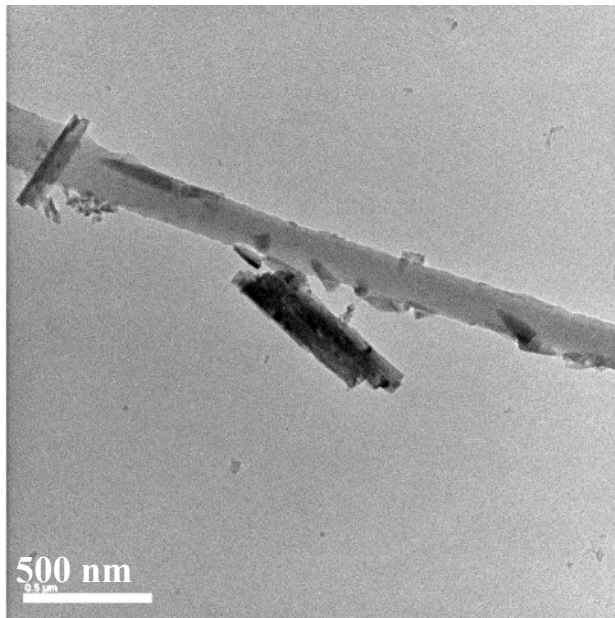
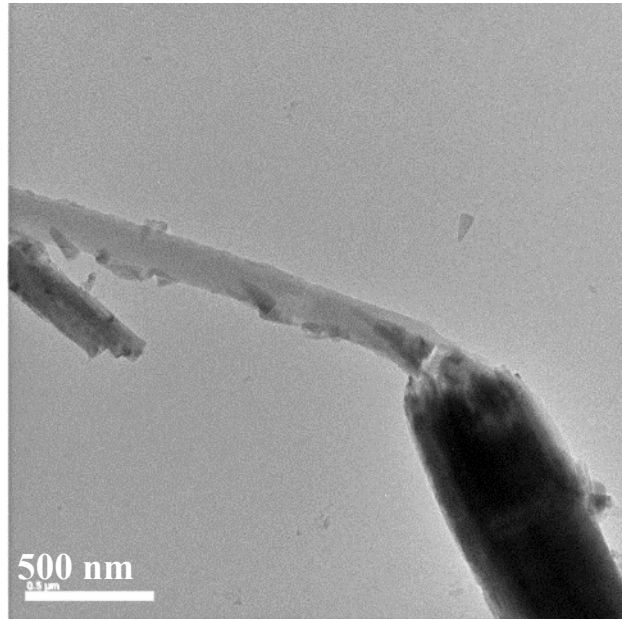
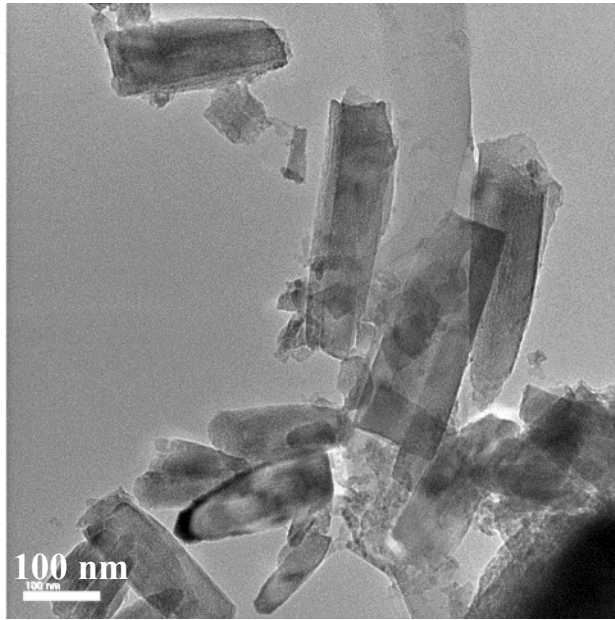
**Supplemental Figure 3. TEM micrographs of tendon mineralized with 27 kDa pAsp for 7 days without fluoride. These fibrils are less highly mineralized as indicated by the diminishing z-contrast that is usually gained from the presence of mineral. In addition, there is a lack of clear striations that indicate the presence of crystals. Some amorphous mineral may be present.**



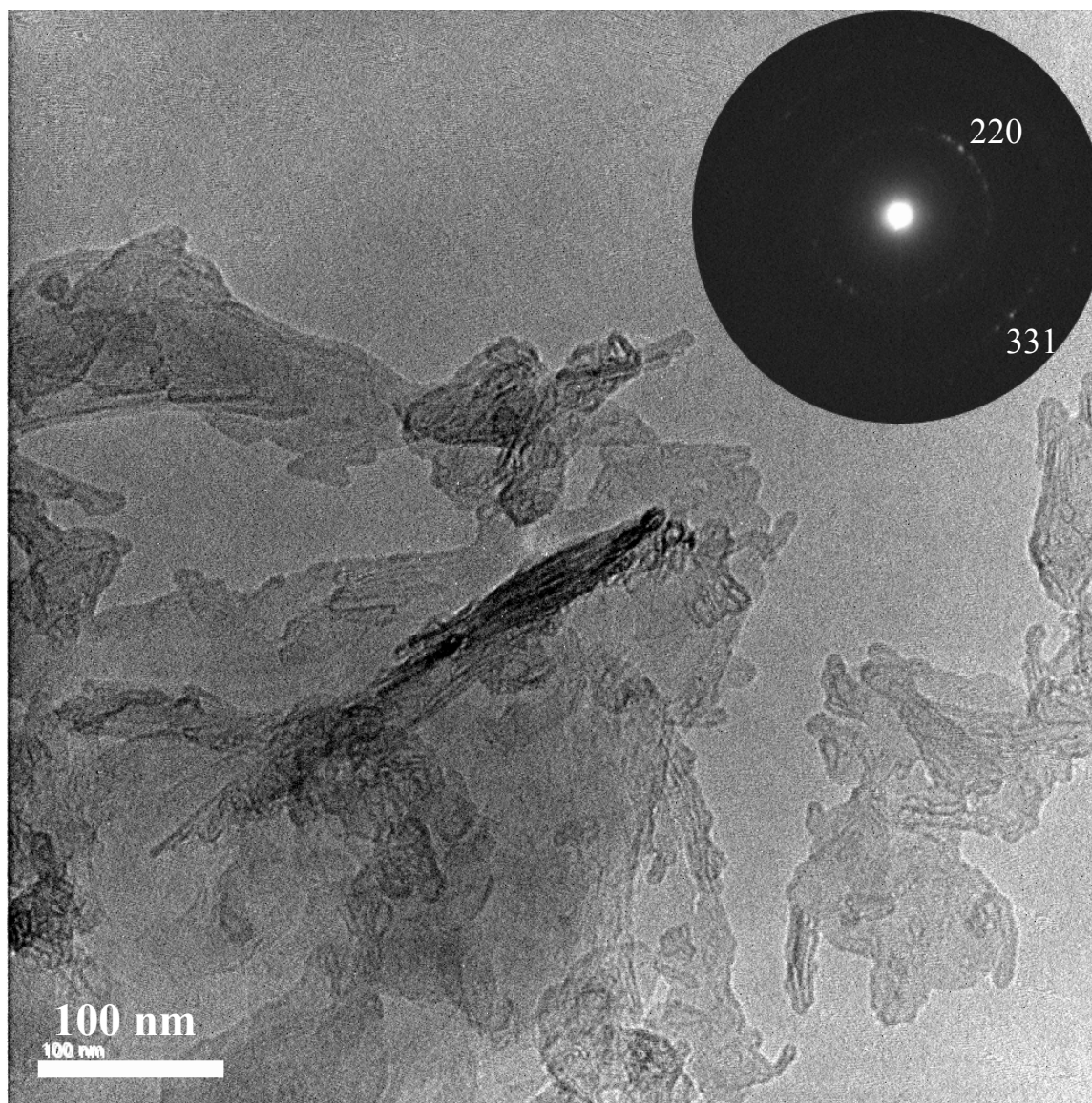
**Supplemental Figure 4. TEM micrographs of tendon mineralized with 27 kDa pAsp for 7 days in the presence of 0.5 ppm F (A) or 1 ppm F (B). A) Tiny, granular crystals not associated with collagen. B) Unmineralized fibrils with a mixture of both granular and rod-like crystals.**



**Supplemental Figure 5. TEM micrographs of tendon mineralized with 27 kDa pAsp for 7 days in the presence of 2 ppm F (A & B) or 5 ppm F (C & D). A) 2 ppm group showing a few fibrils with intrafibrillar mineral that appear to be cemented together. B) 2 ppm group showing poorly mineralized fibrils, with a mix of granular and rod-like crystals that are more randomly dispersed throughout the organic matter (presumably collagen), but with some regions of apparent alignment. C) 5 ppm group showing a fibril fragment with intrafibrillar, aligned apatite crystals surrounded by granular, interfibrillar crystals. D) 5 ppm group showing a mixture of granular and rod-like crystals not associated with collagen.**



**Supplemental Figure 6. TEM micrographs of tendon mineralized with 27 kDa pAsp for 7 days in the presence of 25 ppm F. These fibrils may contain some intrafibrillar mineral (perhaps still amorphous), but primarily have large, extrafibrillar rod-like and/or tiny, granular crystals decorating the surfaces of the fibrils.**



**Supplemental Figure 7. TEM micrograph and SAED pattern of 200 ppm F group, showing large extrafibrillar crystals. SAED (inset) of the crystals displayed spots with d-spacings that most closely match those of calcium fluoride, but at this orientation (without a proper zone axis), the spots cannot be definitively assigned.**