Influence of mineral density and hydration on mineralized collagen fibrils mechanics

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Abstract: Collagen is a ubiquitous protein that possesses incredible mechanical properties. It is highly elastic, shows large strength, and enables large energy dissipation during deformation. Most of the connective tissue in humans consists of collagen fibrils that are composed of a staggered array of tropocollagen molecules and stabilized by intermolecular cross-links. In bone, these collagen fibrils are filled with apatite crystals that play an essential role in the mechanical properties of the bone since they bring stiffness, strength, and wear resistance to enhance the material property of much softer but tougher organic phase.

Using a bottom-up approach, we developed a three-dimensional mesoscale model of collagen fibrils using coarse-grained molecular modeling. We used this model to study the influence of mineral density on the mechanics of the fibrils. We also analyzed role of the hydrated layer present on the surface of the crystals. Mineralized fibrils exhibit a three phase behavior characterized by: (i) an initial elastic deformation corresponding to the collagen molecule uncoiling, (ii) a linear regime dominated by a stick-slip mechanism, and (iii) a stiffening region related to the stretching of the backbone of the tropocollagen molecules combine with mineral collagen sliding. When compared to pure collagen fibrils, mineral significantly improves fibril mechanical properties. The fibril toughness, ultimate strain, and strength to failure can be increased up to 5.8, 6.8 and 8.8 times respectively by adding mineral in the structure. The introduction of a hydrated layer around the crystals leads to decreased adhesion energy between the collagen molecules and the mineral crystal. This results in a significant diminution of the ultimate strength and toughness of the fibrils up to 20 and 50% respectively. On the other hand, hydrated fibrils exhibit a more ductile behavior compared to the catastrophic failure of the fibrils without hydrated layer.

Intrafibrillar mineralization represents a ways to improve intermolecular interaction and lead to an improvement in the tensile strength of collagen fibrils by taking advantage of the full mechanical potential of every collagen molecule in the structure. Minerals improve the intermolecular connectivity by enhancing the interaction between collagen molecules by filling the gap region for collagen assembly. Introducing a hydrated layer improves the ductility of the fibrils, at the cost of losing some strength and toughness.