# Lessons from nature on biomaterial design - structural hierarchy and self-assembly of cartilage biopolymers

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The major components of cartilage extracellular matrix are the fibril-forming collagen and the large proteoglycan aggregates. The organization of the collagen network and the interaction between collagen and proteoglycans has important consequences on the load bearing, transport and diffusion properties of the tissue. Collagen fibers are strong in tension and reinforce the proteoglycan gel. Aggrecan is the major proteoglycan component of the extracellular matrix of cartilage. At physiological concentrations (4-7 % w/w) the high osmotic swelling pressure of aggrecan assemblies, which are enmeshed in the collagen matrix, provides resilience to compressive load, controls lubrication of the joint and protects bone surfaces from wear during articular movement. The structure of the aggrecan bottlebrush (e.g., length of bristles, charge density and distribution) varies with age and state of health. Increased hydration and loss of proteoglycans are the earliest signs of cartilage degeneration during osteoarthritis. To elucidate the differences between healthy cartilage and changes associated with degeneration requires the knowledge of the relationships between molecular and tissue-level properties. The macroscopic properties of cartilage depend on the underlying hierarchical organization at different length scales. Our objective is to gain insight into the biomechanical behavior of cartilage extracellular matrix by a systematic investigation of the physico-chemical properties of proteoglycan assemblies in near physiological salt solution. We report experimental results obtained for the structure and dynamics of cartilage proteoglycans on different length and time scales using osmotic swelling pressure measurements, small angle neutron, small angle X-ray, static and dynamic light scattering and neutron spin echo techniques.