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Mathematics of Biological Charge Transport: Molecules and Beyond

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Cartilage: Hierarchical Organization of Charged Biopolymers

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Understanding the physical and chemical mechanisms affecting cartilage behavior is essential to predict its biomechanical properties, particularly its load-bearing ability, which is governed by osmotic and electrostatic forces that strongly depend on tissue hydration. Such understanding is also important for the success of tissue engineering and regenerative medicine strategies to grow, repair, and reintegrate cartilage. We have developed a multi-scale experimental approach to determine the structure and dynamics of biopolymer assemblies by combining scattering techniques (small-angle X-ray scattering, small-angle neutron scattering, static and dynamic light scattering, neutron spin-echo) with macroscopic methods (osmotic pressure measurements and mechanical measurements). In cartilage the most abundant proteoglycan is aggrecan a large, negatively charged bottlebrush shaped molecule whose complexes with hyaluronic acid provide the osmotic resistance of cartilage, control joint lubrication and protect bone surfaces from wear during articular movement. Osmotic pressure measurements and scattering observations made on aggrecan solutions reveal self-assembly of the bottlebrush molecules into microgels. We found that the structure and dynamic properties of aggrecan assemblies are insensitive to changes in the ionic environment. This behavior is in stark contrast to observations made on highly charged synthetic and biological polyelectrolytes such as poly(acrylic acid) and DNA. The stability of proteoglycan assemblies confers on aggrecan the role of an ion reservoir mediating calcium metabolism in cartilage and bone.