



Macromolecular Nanotechnology

Mechanically strong hyaluronic acid hydrogels with an interpenetrating network structure



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ABSTRACT

Hyaluronic acid (HA) is a natural glycosaminoglycan of high molecular weight with important biological and physicochemical functions. Although hydrogels derived from HA are effective biomaterials for soft tissue regeneration, they are generally brittle, or require complicated synthetic procedures. A simple one-pot synthesis method of mechanically strong HA hydrogels would be attractive for many application areas. Here, we present preparation of HA hydrogels via free-radical copolymerization of methacrylated HA (GMHA) and N,N-dimethylacrylamide (DMA) in aqueous solutions. GMHA was synthesized by methacrylation of native HA at various levels of methacrylation between 4 and 25% corresponding to 115–721 pendant methacrylate groups per GMHA molecule. It was found that GMHA acts as a multifunctional cross-linker during its copolymerization with DMA leading to the formation of interpenetrated and interconnected polymer networks. The effective functionality of GMHA increases with its degree of methacrylation as well as with the DMA concentration. The viscoelastic and mechanical properties of HA hydrogels could be tuned by varying the degree of methacrylation of GMHA and DMA concentration. A significant improvement in the mechanical performance of the hydrogels was observed when DMA is replaced with methacrylic acid monomer. By adjusting the synthesis parameters, hydrogels with a Young's modulus of around 200 kPa could be prepared that sustain up to 20 MPa stresses at 96% compression.

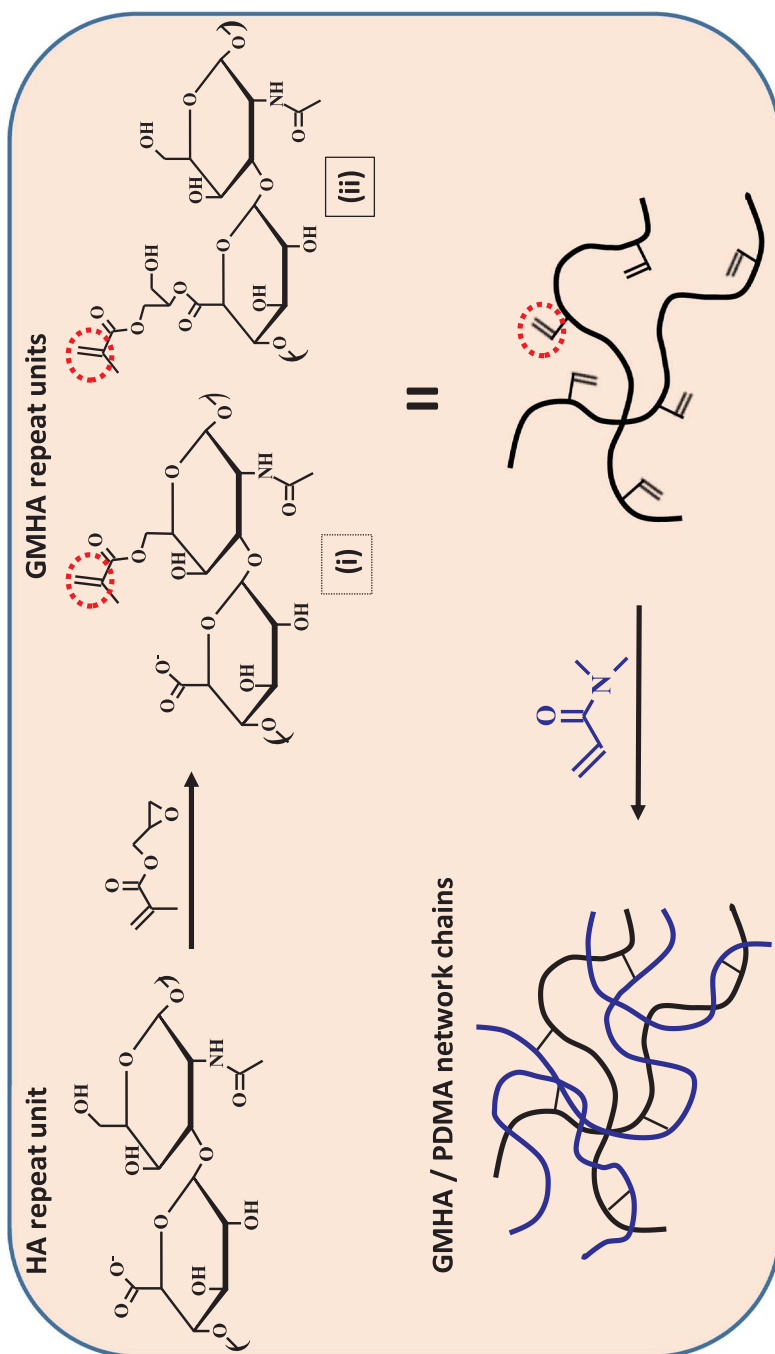
1. Introduction

Hyaluronan or hyaluronic acid (HA) is a natural glycosaminoglycan of high molecular weight composed of disaccharide repeat units of β -1,4-D-glucuronic acid and β -1,3-N-acetyl-D-glucosamine (Scheme 1) [1]. In physiological solutions, HA has a highly extended random coil conformation due to the hydrogen bonding between disaccharide units, and polyanionic properties [2,3]. HA is the major component of the extracellular matrix of vertebrate tissues with important biological and physicochemical functions. Due to the polyelectrolyte nature, HA can absorb large amounts of water and hence acts as lubricant in native extracellular matrix and controls the viscoelasticity of connective tissues [3].

Although HA is an attractive biomaterial for regeneration of soft tissues [4–9], it has limited application areas due to its rapid degradation and poor biomechanical properties. To overcome this drawback, HA has been physically or chemically cross-linked to form HA hydrogels [3,10–12]. The hydrogels based on HA are however generally brittle, or easily dissolve in aqueous solutions. For instance, HA hydrogels prepared by photopolymerization of methacrylated HA in aqueous solutions fracture at around 35% compressive strain under < 60 kPa stresses [13]. Cross-linking of native HA in aqueous solutions using cross-linkers such as divinyl

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Scheme 1. Formation of methacrylated HA (GMHA) by methacrylation of native hyaluronic acid (HA) using glycidyl methacrylate via transesterification (i) and ring opening (ii), and its copolymerization with DMA to form interconnected and interpenetrated GMHA/PDMA network hydrogels. Red circles indicate the methacrylate groups incorporated as pendant into GMHA molecules acting as potential cross-link points during the copolymerization with DMA.

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