Advancing Biobased Chitosan Biomaterials: From Hydrogels to Shape-Recovery Cryogels

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Chitosan-based biomaterials have evolved from composite hydrogels to advanced double-network (DN) systems and shape-recovery cryogels. A central theme across these studies is the use of star-shaped polycaprolactone (stPCL) to overcome the mechanical limitations and poor solubility of traditional chitosan-based materials. Initial research established that star-shaped architectures provide superior segment density and cross-linking efficiency compared to linear homologs, thereby enhancing the stability and mechanical properties of CS/stPCL composite hydrogels fabricated using EDC/NHS conjugation agents.[1] The research progressed into the development of double-network (DN) hydrogels using water-soluble carboxymethyl chitosan (CMCS) and modified poly(ethylene glycol) (PEG), successfully creating a hybrid ionic/covalent system with high shear modulus and bioactivity.[2] Further research led to the fabrication of CS-based networks via urethane/urea linkages, demonstrating that incorporating stPCL significantly improves compressive strength and flexibility while maintaining cell viability above 70%.[3] The most recent development in this work involves chitosan-based cryogels cross-linked through hydrogen-bond interactions with urethane-modified PCL (PCLU). This approach simplifies the fabrication process while introducing rapid shape recovery and a macroporous structure essential for cell infiltration.[4]

References 1) N. Ekapakul, et al., *Polym. Int.* 69, 584-591 (2020). 2) N. Ekapakul, et al., *Carbohydrate Polymers,* 318, 121130 (2023). 3) P. Amonkol, et al., *J. Appl. Polym. Sci.* 141(14), e55191 (2024). 4) S. Phangkam, et al., *ACS Biomater. Sci. Eng.* 11(8), 4747-4757 (2025).