Probing Nonlinear Rheological Behavior of Protein-Pectin Crosslinked Networks via Large Amplitude Oscillatory Shear (LAOS)

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Protein-pectin crosslinked networks are increasingly utilized in food science, biomedical engineering, and soft material design due to their biocompatibility, tunable mechanical properties, and functional versatility. While small amplitude oscillatory shear (SAOS) provides insights into linear viscoelasticity, understanding their nonlinear behavior under large deformations is essential for applications involving processing, mastication, or mechanical loading. In this study, we employ Large Amplitude Oscillatory Shear (LAOS) rheology to systematically investigate the nonlinear viscoelastic response of protein-pectin gels formed via enzymatic (e.g., transglutaminase), covalent (Maillard-induced), and ionic crosslinking mechanisms. By combining Lissajous-Bowditch plots with Fourier-transform (FT) rheology and Chebyshev harmonic decomposition, we resolve the contributions of elastic energy storage (G′) and viscous dissipation (G″) across strain amplitudes (γ_0 = 0.1–500%). Key nonlinear signatures, including strain-stiffening, yielding, and shear-induced network breakdown, are quantified through the framework of intracycle (elastic/viscous nonlinearities) and intercycle (thixotropic/recovery) dynamics. Our results demonstrate that crosslinking density and polymer ratio dictate the transition from linear to nonlinear regimes, with Maillard-crosslinked networks exhibiting pronounced intercycle softening due to irreversible bond rupture, while ionically crosslinked gels show reversible plasticity. Furthermore, we correlate LAOS-derived parameters (e.g., I_3/I_1 ratio, Q_0) with microstructural changes observed via confocal microscopy or scattering techniques. These findings provide a structure-rheology relationship critical for designing protein-pectin composites with tailored mechanical performance in applications such as 3D bioprinting, controlled release systems, or texture-modified foods. The methodology establishes LAOS as a powerful tool for probing the functional limits of soft biological networks under physiologically and industrially relevant deformations.

Keywords: LAOS, protein-pectin gels, nonlinear viscoelasticity, crosslinking, rheology, soft materials, Maillard reaction, harmonic decomposition

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