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

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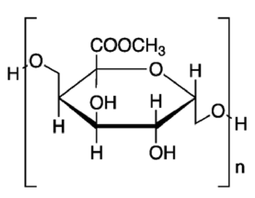
Abstract

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 behaviour 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.

Introduction

Large-amplitude oscillatory shear (LAOS) is a widely used technique for examining the nonlinear viscoelastic behaviour of materials. Unlike linear viscoelastic tests, the resulting stress response is not purely sinusoidal. Instead, the asymmetric stress signal can be expressed as a combination of sinusoidal components, beginning with the fundamental testing frequency and extending to odd multiples of that frequency, known as higher harmonics. Each higher harmonic possesses its own phase angle and provides insights into the material's molecular structure. LAOS data are often represented as parametric stress–strain plots, known as Lissajous–Bowditch curves. These curves typically form S-shaped loops that reflect viscoelastic behaviour, where the loop's width corresponds to the phase angle

Why Pectin?



- It's a carbohydrate that's derived from the pulp and skins of fruits and vegetables (oranges and other citrus fruits, apples, carrots, apricots, plums).
- Pectin is a natural thickener and gelling agent. It's similar to gelatine and often used to make jams and jellies.
- Pectin is rich in soluble fibre, which forms a gel that slows digestion and benefits the health by stabilizing blood sugar and reducing cholesterol levels.

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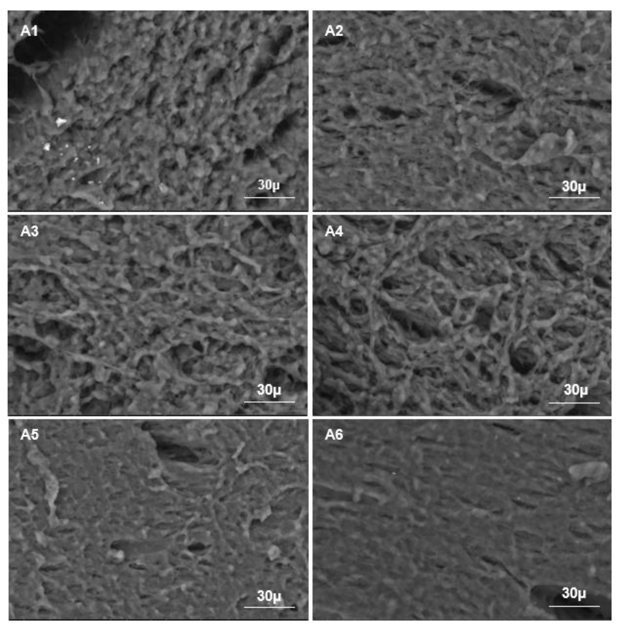


Figure 1. Scanning electron micrographs of extruded composite pea protein-cultured meat cell samples.

Large Amplitude Oscillatory Shear (LAOS)

As two proteins are mixed and heat-induced to form a gel, three interaction scenarios are possible;

(a) Compatibility when both proteins co-assemble into an integrated gel network, enhancing water retention and mechanical strength.

(b) Semi-compatibility when one protein forms the primary gel network, while the other interacts with it, modifying rigidity and elasticity. This often increases hardness and water retention, though effects are protein dependent.

(c) Incompatibility when proteins form separate networks or distinct gel domains, leading to phase separation, weakened mechanical properties, and heterogeneous microstructure.

Nonlinear viscoelastic properties of the extrudates were evaluated using a Discovery HR-20 rheometer (TA Instruments, New Castle, DE, USA) equipped with a 40-mm parallel plate cross-hatched geometry. Samples were equilibrated at 20 °C for 10 min before testing. The samples were subjected to a 2.5 N fixed normal force and transient sinusoidal deformation at 22 different strain levels ranging from 0.01% to 100% at a frequency of 1 Hz.

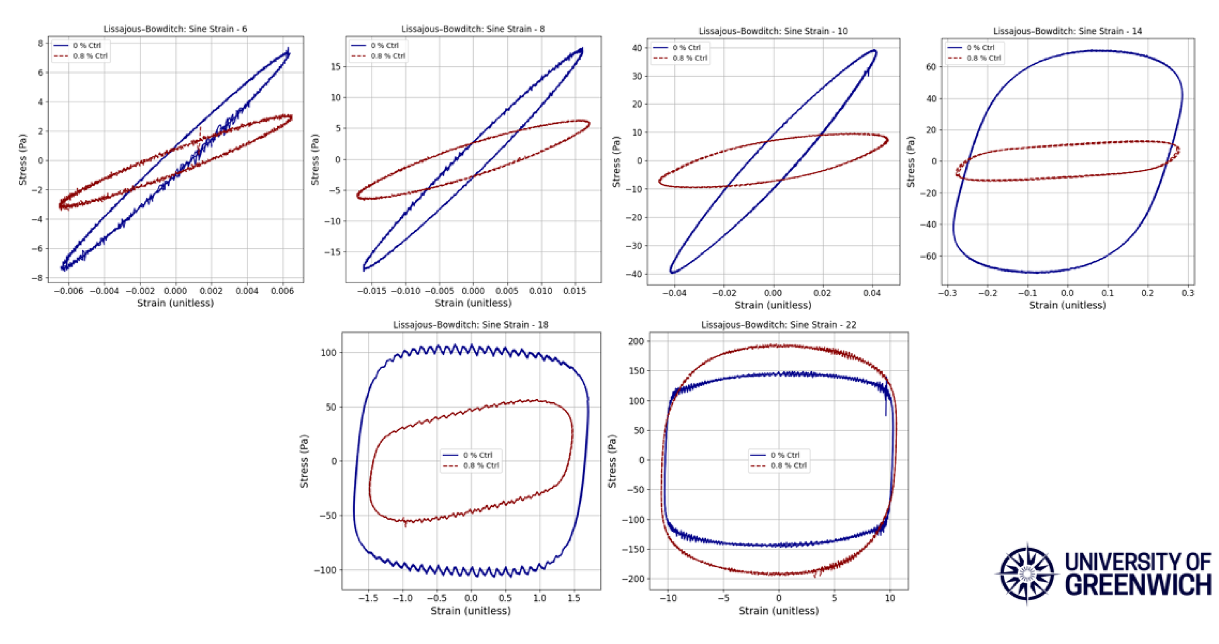


Figure 2. Effect of pectin only on the LAOS data

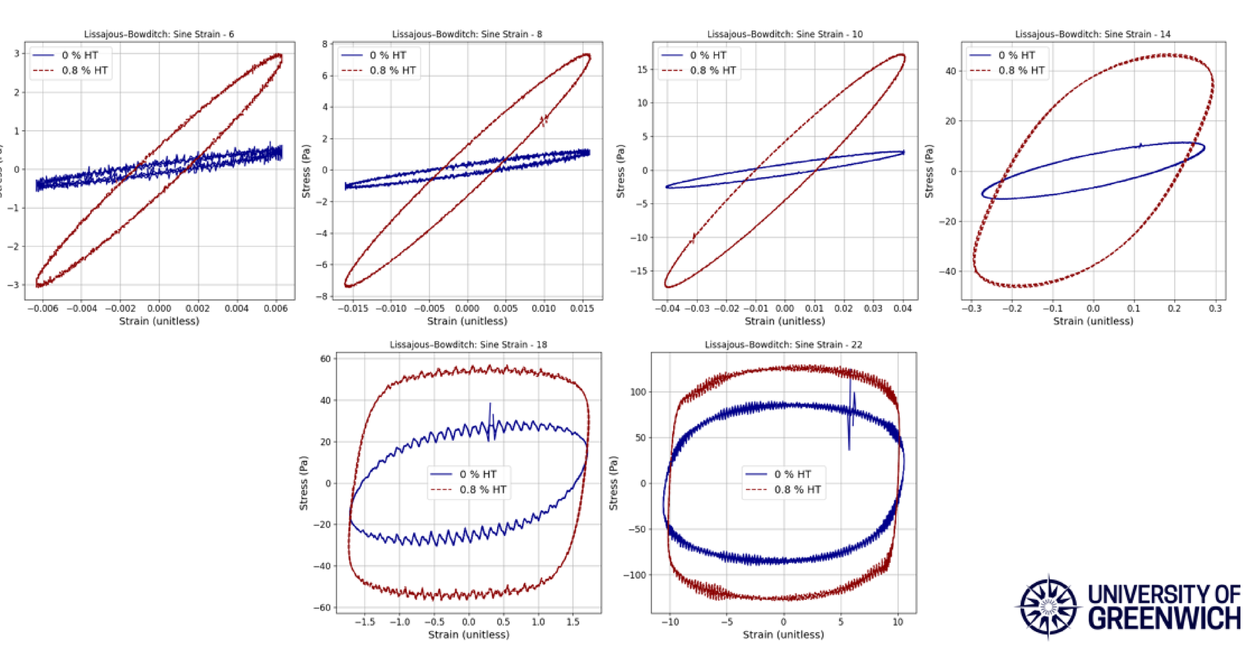
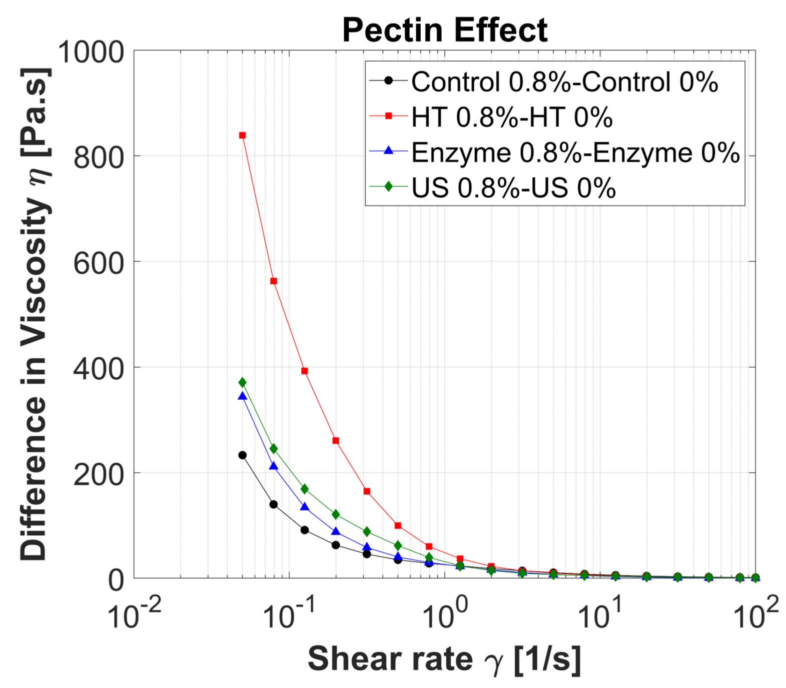


Figure 3. Effect of pectin and heat treatment on the LAOS data

Results



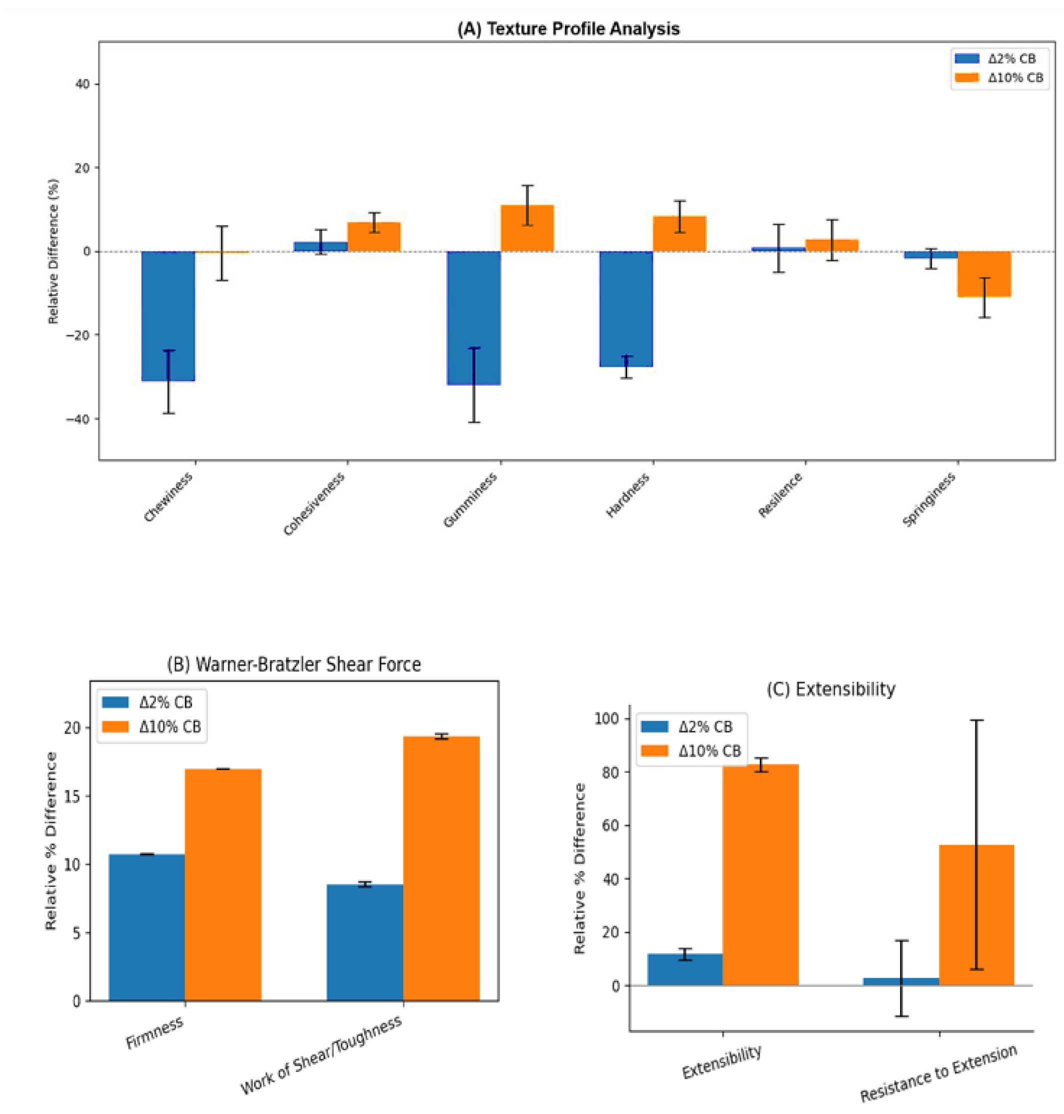


Figure 4. (A) Textural properties obtained from the Texture Profile Analysis (TPA), (B) Warner-Bratzler (WB) Shear Force (Meat Tenderness), (C) Extensibility

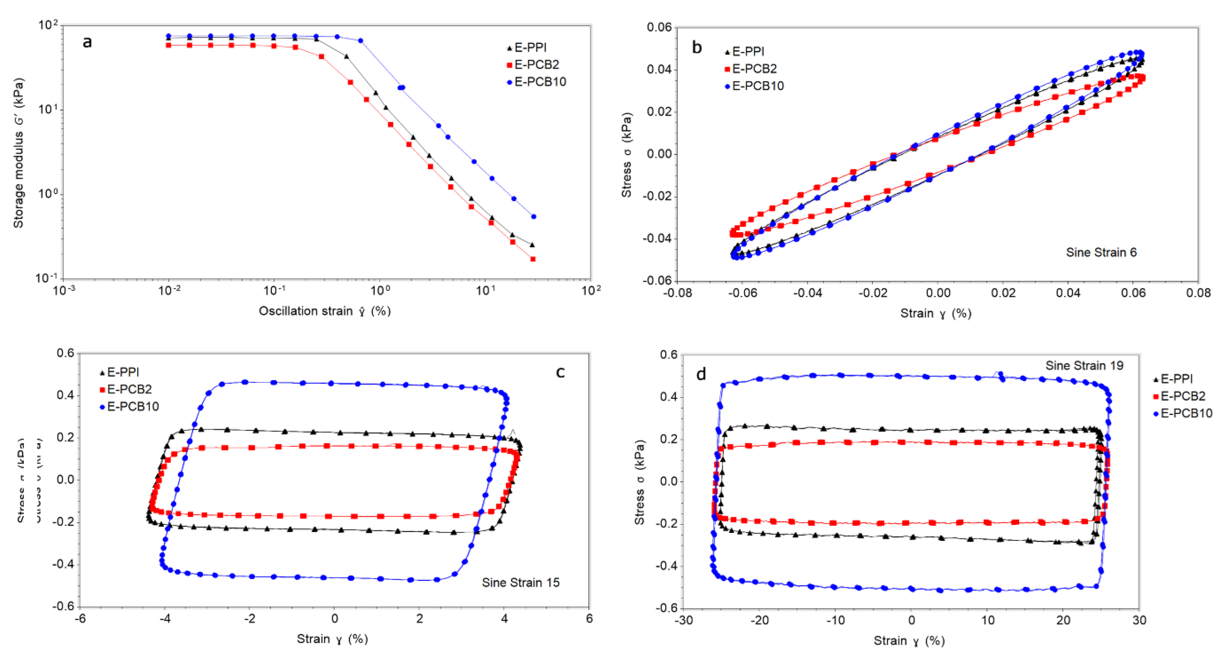


Figure 5. Amplitude sweep (a) and Large Amplitude Oscillatory Shear (LAOS) Lissajous–Bowditch curves (b-d).

Future work:

- Apply same technique to potato starch extrudates
- Train for machine learning modelling
- Apply it to protein with fibre samples

References

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