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Analysis of Pectin Biopolymer Phase States using Acoustic Emissions

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Abstract

Acoustic emissions are stress or elastic waves produced by a material under external load. Since acoustic emissions are generated from within and transmitted through the substance, the acoustic signature provides insights into the physical and mechanical properties of the material. In this report, we used a constant velocity probe with force and acoustic emission monitoring to investigate the properties of glass phase and gel phase pectin films. In the gel phase films, a constant velocity uniaxial load produced periodic premonitory acoustic emissions with coincident force variations (saw-tooth pattern). SEM images of the gel phase microarchitecture indicated the presence of slip planes. In contrast, the glass phase films demonstrated early acoustic emissions, but effectively no force or acoustic evidence of periodic or premonitory emissions. Microstructural imaging of the glass phase films indicated the presence of early microcracks as well as dense polymerization of the pectin (without evidence of slip planes). We conclude that the water content in the pectin films contributes to not only the physical properties of the films, but also the stick-slip motion observed with constant uniaxial load. Further, acoustic emissions provide a sensitive and practical measure of this mechanical behavior.

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Keywords

acoustic emissions; pectin; Scanning electron microscopy

Introduction

Acoustic emissions are defined as stress or elastic waves produced by a material under external load (Skal's'kyi, Andreikiv, & Serhienko, 2003). Stored energy, rapidly released from within a material, can produce a wide range of acoustic signals (Kohn, 1995; Maslov & Gradov, 1986). The acoustic emissions can vary from a large number of low amplitude acoustic events to relatively few acoustic events of large amplitude (Bartenev & Fadeev, 1991; Maslov & Gradov, 1986; Ravi & Sethuramiah, 1995). Since acoustic emissions are generated from within and transmitted through the substance, the acoustic signature provides insights into the physical and mechanical properties of the material.

Previous acoustic emission studies have predominantly focused on inorganic hard solid objects such as metals and rocks (Bartenev & Fadeev, 1991; Skal's'kyi et al., 2003), but many acoustic studies have provided insights into organic solids and soft objects including plants (Vergeynst, Sause, De Baerdemaeker, De Roo, & Steppe, 2016). Acoustic emissions in plants have focused on the sounds and forces associated with the crushing of foodstuffs. Acoustic emissions have been studied during mechanical destruction of fruit (Taniwaki, Hanada, & Sakurai, 2009; Zdunek, Cybulska, Konopacka, & Rutkowski, 2011), baked goods (Chen, Karlsson, & Povey, 2005), and vegetables (Alvarez, Saunders, Vincent, & Jeronimidis, 2000; Zdunek, 2012). These studies have provided an objective assessment of food texture (Drake, 1963; Luyten, Pluter, & van Vliet, 2004). An interesting challenge has been linking these acoustic properties to subjective assessments of food freshness and wholesomeness (Christensen & Vickers, 1981; Fillion & Kilcast, 2002; Vickers, 1982, 1984).

More recently, acoustic emission studies have been applied to hydrogels (Scognamiglio et al., 2018), and human tissue (Urban, Silva, Fatemi, & Greenleaf, 2006). A particularly interesting hydrogel is the heteropolysaccharide pectin. Predominantly composed of partially esterified linear chains of (1,4)-alpha-D-galacturonic acid residues (Monsoor, Kalapathy, & Proctor, 2001; Nunes et al., 2012), pectin comprises approximately 30% of the primary cell walls of many plants (Scheller, Jensen, Sorensen, Harholt, & Geshi, 2007). Also, pectin has been shown to bind to the surface of visceral organs---a finding with implications for a variety of biomedical application (Servais, Kienzle, Valenzuela, et al., 2018; Servais, Kienzle, Ysasi, et al., 2018). Relevant to biomedical applications are the striking and reversible physical properties of pectin induced by small changes in water content (Einhorn-Stoll, 2018; Iijima, Nakamura, Hatakeyama, & Hatakeyama, 2000; Panchev, Slavov, Nikolova, & Kovacheva, 2010). In pectins with a high degree of methyl esterification, the loss of water can produce a glass-like polymer that is hard and brittle. In contrast, an increase in hydration can produce a polymer that is soft and rubbery. Currently, there are limited nondestructive means of distinguishing glass and gel phase films. Whether acoustic emissions can distinguish between these pectin phase states is unknown.

In this report, our goal was to assess the utility of acoustic emissions in distinguishing between glass phase and gel phase pectin films. We monitored the acoustic emissions of various pectin biopolymers under a constant velocity uniaxial load. The acoustic signatures of these phase states were correlated with mechanical properties and microstructural imaging of the pectin biopolymers.

Methods

Pectin.

The unstandardized high-methoxyl citrus pectins were obtained from a commercial source (Cargill, Minneapolis, MN, USA) with a glycosyl residue content of 78–86% mole % galacturonic acid, 2–3% rhamnose, 9–14% galactose and 0.5–5% arabinose based on gas chromatography–mass spectrometry of trimethylsilyl derivatives (Biswal et al., 2017) and with a 77–85% homogalacturonan and 16–23% RGI content. The proportion of galacturonic acid residues in the methyl ester form determined the degree of methoxylation. High-methoxyl pectins (HMP) were defined as those pectin polymers with a greater than 50% degree of methoxylation (Mean=67±8%). The pectin powder was stored in low humidity at 25°C.

Pectin dissolution in water.

The pectin powder was dissolved at 25°C by a step-wise increase in added water to avoid undissolved powder and aggregates (Panchev et al., 2010). Exogenous heat was not used. Swelling and softening of the particles was followed by fluidization and dissolution (Furmaniak, Terzyk, & Gauden, 2007). Pectin titrations determined optimal concentrations for the dissolution of the pectin (3% w/w). Mixing was achieved at 25°C by a high-shear 10,000 rpm rotor-stator mixer (L5M-A, Silverson, East Longmeadow, MA USA). RPM peak and plateau viscosity was monitored using digital tachometer and ammeter (DataLogger, Silverson). The dissolved pectin was poured into a standard mold for further studies. The pectin film thickness was either 50µm or 100µm.

Humidification chamber.

The hydration of the pectin biopolymers was controlled using humidification chambers. The humidity chambers were custom designed 5.7-liter translucent polycarbonate humidification. Humidification was produced by an ultrasonic humidifier or manual aerosol device. The chamber was monitored by wireless (Bluetooth) hygrometer and thermometer sensors (Inkbird, Shenzhen, PRC). The data recording device was maintained within the humidification chamber throughout each experiment. Glass phase (6–13%) and gel phase (25–31%) pectins, consistent with their use in biomedical applications, were the focus of these studies.

Fracture mechanics.

To elicit acoustic emissions, the pectin biopolymers were subjected to a controlled uniaxial load normal to the plane of the polymer film using an TA-XT plus (Stable Micro Systems). After calibration of the 5 kg load cell, a 5 mm stainless steel spherical probe was mounted to a 5 kg load cell and positioned centrally over the biopolymer. The test fixture was comprised

of a 25mm circular template that secured the biopolymer but did not hinder probe penetration and rupture. The tests were preprogrammed for reproducibility. The probe descended at a pre-test speed of 1 mm/sec. A trigger force of 5g/0049N was used to detect the polymer surface; the trigger force initiated the burst protocol. To burst the film, the probe compressed the films at a test speed of 2 mm/sec until rupture; rupture was reflected in the sudden drop of the compression force. The fracture force and distance, synchronized with the acoustic recordings, were recorded at 500 pps. In the analysis of probe diameter, larger probes were associated with an increase in acoustic intensity, but an identical temporal pattern.

Sound recordings and analysis.

Sound was detected by a cardoid polar pattern condenser microphone (AT2020, Audio-technica, Stow, OH, USA) with a dynamic range of 20–20,000 Hz and a low-mass diaphragm for effective transient response. The sound was processed by a U-Phoria UMC22 audio interface with Xenyx microphone preamplifier (Behringer, Willich, Germany). The data was stored as uncompressed WAV audio format files. Audio processing, editing and analysis was performed with Audacity 2.3.0 (open source), Wavesurfer (open source) and custom acoustic software (MatLab, MathWorks, Natick MA, USA).

Spectral analysis.

The acoustic spectral analysis transformed the time-domain digital recording into a frequency domain spectrum using the fast Fourier transform (FFT) function in MatLab (MathWorks Natick, MA, USA). The spectral analysis was plotted as a graph of frequency (in Hz) versus intensity (dB). The frequency ranged from 2 Hz to 24000 Hz. The FFT signal was displayed visually with degrees of amplitude represented along a light to dark dimension (white=no energy; dark=increased energy). The Fourier analysis window was calculated based on a wide band spectrogram.

Acoustic power.

Acoustic power was defined as the rate at which recorded acoustic energy was detected per unit time from the polymer source. The acoustic field and recording parameters were kept constant. Because of the absence of an absolute calibration standard, the integration of the power plots was expressed as a relative power index.

Scanning electron microscopy.

After coating with 20–25 Å gold in an argon atmosphere, the pectin films were imaged using a Philips XL30 ESEM scanning electron microscope (Philips, Eindhoven, Netherlands) at 15 Kev and 21 µA. Stereo pair images were obtained using a tilt angle difference of 6° on a eucentric sample holder using standardized automation.

MicroCT scanning.

The microCT scans were performed on pectin polymers subjected to burst testing. The films were imaged using a SKYSCAN 1272 microCT scanner. The scanner obtained up to 209 Megapixel (14450×14450 pixels) virtual slices through the films, more than 2600 such slices

after a single scan using a 16-megapixel camera in triple offset mode. Using phase-contrast enhancement, the scanner was sensitive to structural details as small as 0.35 μm .

Statistical analysis.

The unpaired Student's *t* test for samples of unequal variances was used to calculate statistical significance. The data was expressed as mean $\pm$ one standard deviation. The significance level for the sample distribution was defined as $p < .01$. Software analysis was performed by R software (www.R-project.org) and Microsoft Excel 365 analysis tools (Redmond, WA, USA).

Results

Acoustic emissions in pectin phases.

To compare the acoustic signature of pectin biopolymers with distinct physical properties, films were prepared with water content characteristic of both the glass phase (6–13% w/w) and gel phase (25–31% w/w). The films were tested for their response to a constant velocity uniaxial load applied normal to the plane of the film. In the glass phase films, probe contact with the films resulted demonstrated variable early acoustic emissions (Figure 1A, black arrow); subsequent acoustic emissions were absent until film rupture (Figure 1A, gray arrow). In contrast, initial probe contact with the gel phase films demonstrated no early acoustic emissions. With progressive deformation of the film, the gel phase films demonstrated detectable force fluctuations (saw-tooth pattern) prior to film failure (Figure 1B, bracket). Coincident with the force fluctuations, the gel phase films demonstrated a periodic acoustic emission prior to film rupture (Figure 1B, ellipse).

Microcracks and burst emissions.

To provide more detailed analyses of these observations, acoustic spectral analysis was performed. The pectin film acoustic emissions were analyzed pre-rupture (Figure 2A) and during film rupture (Figure 2B). In comparing pre-rupture emissions, the presumed microcracks in the glass phase films were associated with greater intensity emissions in the higher frequencies when compared to the acoustic spectra of the pre-rupture gel phase films (Figure 2A, bracket). This tendency toward higher frequency emissions in the glass phase films was also noted in the rupture (fracture) emissions (Figure 2B). The acoustic intensity of the burst fracture of both the glass and gel phase films was greater than pre-rupture (premonitory) emissions (Figure 2B).

Similar to the pre-rupture emissions, the glass phase films demonstrated greater intensity emissions in the higher frequencies than the gel phase films (Figure 2B, bracket). MicroCT analysis of fractured glass phase pectin films demonstrated evidence of both macrocracks (Figure 3A) and microcracks (Figure 3B).

Acoustic power.

Acoustic power plots, reflecting sound energy as a function of time, also demonstrated a clear distinction between glass phase and gel phase pectin films. Again, the glass phase films produced sporadic pre-rupture emissions interpreted as microcracks (Figure 4A, arrows). In

contrast, the gel phase films produced multiple periodic emissions reflected in the power plot (Figure 4B). The integration of the power plots, expressed by a relative power index, demonstrated significantly more acoustic power produced in the gel phase (0.51 ± 0.18) compared to the glass phase films (0.32 ± 0.14) ($p = .027$; $N = 50$ films). Notably, the acoustic power of the rupture event itself was not different between gel phase and glass phase films ($p > 0.4$).

Acoustic intervals.

In gel phase films, the periodicity of the acoustic interval reflected two characteristic patterns (Figure 5). Figure 5 The films demonstrated either 1) a relatively similar intensity pattern or 2) a pattern demonstrating both a high intensity and low intensity acoustic signal. In gel phase films with relatively similar acoustic intensity, the acoustic interval was regular, with evidence of a decreasing time interval near film rupture (Figure 5A). In films with a dual intensity pattern, the interval between high intensity emissions was nearly 3.89 ± 1.1 times longer than the lower intensity emissions (Figure 5B) ($p < .001$; $N = 20$ films) with a proportionate increase in signal intensity (4.4 ± 0.5 -fold).

Film microstructure.

To assess the structural changes associated with the acoustic emissions, we studied both glass and gel pectin films by SEM. A comparison of gel and glass phase microstructure was performed by SEM. Glass phase films demonstrated near-complete polymerization of the pectin (Figure 6A). In contrast, gel phase films demonstrated areas of indistinct microstructure associated with potential slip interfaces (Figure 6B, white arrows) and amorphous spaces (Figure 6B, black arrow).

Discussion

In this report, our goal was to investigate the utility of acoustic monitoring in distinguishing between glass phase and gel phase pectin films. To investigate the different pectin phase states, we used a constant velocity probe with force and acoustic emission monitoring. In the gel phase films, a constant velocity uniaxial load produced periodic premonitory acoustic emissions with coincident force variations (saw-tooth pattern). SEM images of the gel phase microarchitecture indicated the presence of slip planes. In contrast, the glass phase films demonstrated early acoustic emissions, but effectively no acoustic evidence of periodic or premonitory emissions. Microstructural imaging of the glass phase films indicated the presence of early microcracks as well as near-complete polymerization of the pectin (without evidence of slip planes). We conclude that the water content in the pectin films contributes to not only the physical properties of the films, but also the stick-slip motion observed with constant uniaxial load. Further, acoustic emissions provide a sensitive measure of the phase state (water content) of pectin films and a practical assessment of their mechanical behavior.

In our studies, water content of 25–31% (gel phase) and water content of 6–13% (glass phase) were associated with striking differences in film properties. 1) Glass phase films were hard and brittle, whereas gel phase films were soft and rubbery. 2) Gel phase films

demonstrated significantly greater extensibility, or strain extension, than the glass phase films. 3) Gel phase films were associated with periodic acoustic emissions and stick-slip motion, whereas glass phase films were not. These findings underscore the importance of water content in the mechanical properties of pectin films.

Pectin content, in contrast, determined the rupture strength of the films. Pectin film burst or rupture strength was independent of water content between 6 and 31%. Similarly, acoustic power plots of film rupture were independent of water content. Finally, burst strength was independent of loading velocity (not shown). These results indicate that the amount of pectin, and not the films' water content, determined the energy required for film rupture.

The saw-tooth force pattern and the periodic acoustic emissions in the gel phase pectin films are consistent with a repetitive process of molecular sticking and slipping. Juddering or jerky movements are a common experience in physical and biologic systems. Also known as friction-induced vibration or stick-slip motion (Berman, Ducker, & Israelachvili, 1996; Ibrahim, 1994), this phenomenon is a result of two rubbing surfaces responding to a constant input force or velocity (Rice & Ruina, 1983). In the simplest explanation, there is high static friction in the glass phase because the pectin has near-completely solidified. The probe force can exceed the static friction only at the point of film rupture. In the gel phase films, SEM imaging suggests that the increased water content lowers the density of pectin polymerization and facilitates the formation of slip planes—structural changes likely associated with lower static friction (Hull, 1999; Lawn & Marshall, 1994). Consequently, the kinetic friction of the gel phase system can intermittently exceed this value resulting in the periodic stick-slip motion observed here. An interesting finding in this study was that acoustic emissions were dependent upon the water content of the polymer. The pectin polymers with a water content of greater than 25% (w/w) consistently demonstrated pre-rupture acoustic emissions. Although there were variations in the amplitude of acoustic emissions, almost all gel phase emissions were periodic and premonitory. In extrapolating our experiments, the gel phase acoustic intervals combined with the known probe velocity of 2mm/sec, suggests that the average slip distance was approximately 80 μm . This intriguing observation suggests the possibility that pectin variants may demonstrate distinctive stick-slip motion.

A limitation of this study is that we focused on the effect of water content on the acoustic emissions of only one naturally occurring variant of pectin---namely, high-methoxyl citrus pectin. We speculate that acoustic emissions will be a useful tool to explore the mechanical and structural properties of all naturally-occurring polymers. We anticipate that complementary measures of this motion, such as vibrometry and accelerometry, may be useful in these studies.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

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Abbreviations:

HMP	high-methoxyl pectin
SEM	scanning electron microscopy
Wc	water content

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Highlights

- A uniaxial load applied to high-methoxyl pectins produces a distinctive acoustic signature
- Glass phase films produce isolated emissions prior to rupture
- Gel phase films produce periodic emissions prior to rupture
- Periodic emissions correlate with slip planes seen on scanning electron microscopy

Figure 1.

Acoustic emissions associated with burst testing of glass phase and gel phase pectin films.

The burst test was performed at a standard probe velocity (2 mm/sec) and acoustic environment. Force variation is shown in the upper panel. Corresponding time-domain responses (Waveform) and frequency spectra (Spectrogram) are shown in lower panels.

Spectrogram amplitude is represented by grayscale intensity (gray<black). A) Glass phase films, defined by 7% water content, demonstrated acoustic emissions early after probe contact (black arrow). B) Gel phase film, defined by 20% water content, demonstrated no early acoustic emissions, but periodic pre-rupture emissions. The spectrogram demonstrated increasing amplitude in premonitory emissions (ellipse). Representative burst tests of glass phase and gel phase films are shown. An image of the burst probe and fixture is shown (A, inset).

Figure 2. Spectral density estimation of pectin acoustic emissions pre-rupture (A) and during film rupture (B). A) The pre-rupture emissions were recorded in representative films from the glass phase (10%, gray) and gel phase (26%, black). A comparison of the spectra demonstrated a consistent difference at the higher frequencies (bracket). B) The acoustic emissions of the rupture (fracture) of glass phase (10%, gray) and gel phase (20%, black) films are shown (0.1 seconds). A significant difference in the fracture spectra was again noted in the higher frequencies (bracket).

Figure 3. MicroCT images of a fractured glass phase pectin film. In addition to the macrocracks typical of fractured glass phase films (A, arrows), linear microcracks were commonly detected (B, arrows). Similar microcracks were detected in many, but not all, films with early acoustic emissions. The occasional negative results potentially reflects limitations in detection sensitivity.

Figure 4.

Acoustic power plots, reflecting acoustic energy per unit time, were calculated for glass phase and gel phase films undergoing burst testing. A) Glass phase films (6–13% water content) demonstrated sporadic early peaks (arrows). B) Gel phase films (25–31% water content) demonstrated periodic peaks leading to film rupture (ellipse). Power plots represent four randomly selected burst tests.

Figure 5.

Time-domain response patterns of gel phase acoustic emissions. A) Relatively regular pattern of acoustic emissions. The mean interval between emissions was $.04 \pm .01$ sec (range 0.02 to 0.07 sec) with increasing variability prior to rupture. B) Acoustic emissions that showed dual intensity waveforms demonstrated a correlation between signal interval and signal intensity. The interval between high intensity emissions was 3.8-fold longer than lower intensity emissions. Similarly, there was a 4.4-fold increase in signal intensity. Representative films are shown.

Figure 6. Scanning electron microscopy images of pectin films. A) SEM images of a glass phase pectin film. The pectin appears to be near-solid. B) SEM image of a gel phase pectin film with apparent slip planes (white arrows) and amorphous spaces (black arrow) between pectin polymers.