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Dependence of concentration and heating-cooling rate on rheological properties of *Plantago lanceolata* seed mucilage

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Abstract

In this study, the effect of concentration (0.5, 1, 1.5 and 2%) and heating-cooling rate (1, 5 and 10 °C.min⁻¹) on the rheological properties of *Plantago lanceolata* seed mucilage (PLSM) solutions were investigated. It was observed that the gum dispersions exhibited viscoelastic properties under the given conditions. Mechanical spectra of PLSM were classified as weak gels based on the frequency sweep, complex viscosity (η^*) and $\tan \delta$ results. All variables had significant impacts on the rheological parameters. Chemical and monosaccharide compositions were also determined to provide more structural information. The results revealed that PLSM had high total sugar content (87.35%), and it is likely an arabinoxylomannan-type polysaccharide.

Keywords: *Plantago lanceolata* seed mucilage; Rheology; Sugar composition.

1. Introduction

Hydrocolloids, also called gums, are macromolecules that dissolve or disperse in water and form viscous dispersions or gels [1]. Hydrocolloids play a great role in food and pharmaceutical industries due to their wide array of functions including; thickening, gel forming, texture producing, emulsifying, stabilizing, controlling crystallization and preventing syneresis [2]. The most important hydrocolloids in food systems are pectin,

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galactomannans, carrageenan, alginate, agar, Arabic gum, starch and cellulose and their derivatives [3].

These polymers have the advantage of being considered as totally natural for many consumers [4]. Selection of a suitable hydrocolloid for a system depends on hydrocolloid's functions and desirable properties in foods [5], aside from their costs and safety [6]. The annual use of hydrocolloids is constantly increasing and consequently recent efforts have been focused on finding new sources of more efficient and economic hydrocolloids.

The rheological behavior of hydrocolloids is of special importance when they are used for modifying textural attributes. It is also well-recognized that rheological properties play a vital role in process design, evaluation and modeling and are considered as measurement for product quality (e.g. indication of total solids or change in molecular size). Rheological data is required for calculation in any processes involving fluid flow (e.g. pump sizing, extraction, filtration, extrusion, purification) and plays an important role in the analyses of flow conditions in food processes such as pasteurization, evaporation, drying and aseptic processing [7].

Dynamic and steady shear rheological properties can be used to provide insight on the structural properties of a sample [8]. Small-amplitude oscillatory shear (SAOS) measurements are a type of dynamic rheological test in which, stress and strain are varied harmonically with time in the linear viscoelastic region (LVR). Expanding the database on the viscoelastic properties of gum solutions/dispersions is critical in food processing in order to adjust processing parameters, monitor consistency as well as predicting the stability of fluid food systems and the final textural attributes of formulated food. Therefore, dynamic rheology is one of the methods used most extensively to assess the viscoelastic behavior of polysaccharide solutions/dispersions or gels. The viscoelastic properties of various gums such as xanthan, guar [9], pectin [10], *Opuntia ficus indica* mucilage [11], basil seed gum [12], gum tragacanth [13], *Lepidium perfoliatum* seed gum [14] and *Alyssum homolocarpum* seed gum [1] have been reported by other researchers.

Generally, the rheological properties of hydrocolloid solutions are influenced by many parameters such as concentration of the active compound, temperature, degree of dispersion, dissolution, electrical charge, previous thermal and mechanical treatment, presence or absence of other lyophilic colloids, the presence of electrolytes and non-electrolytes, pH, ionic strength and salts [3,15].

Plantago, of the plantaginaceae family, is one of the most plentiful and widely distributed medicinal plants throughout the world. *Plantago lanceolata* L., also known as Ribwort or Ribwort Plantain [16,17], produces large amounts of seeds. These seeds have for a long time been used as an anti-catarrhal, respiratory demulcent [16], subtle antibiotic, cough and bronchitis remedies [18]. *Plantago lanceolata* seed mucilage (PLSM) is extracted predominantly from the outermost layer of the seed hull. The hull release mucilaginous substance when immersed in water. Accordingly, aqueous extraction is one of the most popular techniques used for the extraction of seed mucilage. The rheological properties of PLSM are of high interest in world due to its widespread usage in the traditional pharmaceutical prescriptions [19].

As a result of diversity in different gum structures and extrinsic conditions within the fluid food systems, the rheological behaviors are different from one gum solution to another [14]. Hence, food companies may have a hard time deciding which gums to use in their fluid food formulations. Thus, an understanding of the rheological properties of PLSM is essential for evaluating its potential applications and use as food thickeners or stabilizers.

However, despite the great potential of temperature impact on rheological properties of this rarely investigated hydrocolloid, no extensive research has been performed on the rheological properties of PLSM when commonly used thermal processes. There is no published information about the viscoelastic behaviour of this mucilage. In this study, the objectives were to investigate dynamic rheological properties of PLSM and its dependency to concentration and temperature. The effect of consecutive heating–cooling cycles on the viscoelasticity was also determined.

2. Materials and methods

2.1. Materials

P. lanceolata seeds were supplied by the local market in Mashhad, Iran. The cleaned *P. lanceolata* seeds were wrapped in plastic bags, sealed and kept in dry and cool place. All chemicals used were of analytical grade.

2.2. Mucilage extraction

Husks of seeds were dispersed in distilled water at a water/husk ratio of 20:1. The husk–water slurry was stirred continuously with a mechanical mixing paddle throughout the entire extraction period (1 h). Husks of *P. lanceolata* seeds were separated from the seed coats by

mechanical milling of the outer layer of the seeds using a 27-cm basket centrifuge lined with a 1-mm mesh. The seeds were discarded and ultimately, the dispersion was dried in a conventional oven (overnight at 45 °C), milled and sieved using a mesh 18 sifter [20]. The powder was stored in an air-tight test tube at room temperature for further experiments.

2.3. Chemical analysis

In order to determine moisture, fat, ash and protein contents of PLSM samples, AOAC method was used [21]. The protein content was quantified using nitrogen to protein conversion factor of 6.25. Total carbohydrate content was subtracted from 100% – (Moisture + Ash + Fat + Protein).

The monosaccharide analysis was performed by NREL two-step acid hydrolysis and quantified using HPAEC-PAD. In brief, ca. 30 mg dry matter PLSM material was mixed with 72% H₂SO₄ and left for 1 h to react at 30 °C. The reaction mixture was then diluted to 4% H₂SO₄ and hydrolyzed in an autoclave at 120 °C for 40 min. [22]. HPAEC separation of the PLSM was performed using a HPAEC-PAD, ICS5000 system (Dionex Corp. Sunnyvale, CA) equipped with a CarboPac™ PA1 column by a method principally as described by Arnous and Meyer [23]. L-arabinose, L-rhamnose, D-galactose, D-glucose, D-xylose, D-mannose, D-galacturonic acid, and D-glucuronic acid were used as monosaccharide standards for quantification, which was performed using Chromeleon software (Dionex Corp. Sunnyvale, CA). Recovery values for the monosaccharides were estimated from parallel runs of monosaccharide standards.

2.2. Samples preparation

After preparing dried mucilage, aqueous dispersions of PLSM were prepared at different concentrations (0.5, 1, 1.5 and 2% w/v) in deionized water. Different concentrations of NaCl and CaCl₂ (50, 100, 500 and 1000 mM) were added separately at every concentration of PLSM while stirring for 30 min. Finally, 10 ml of dispersions were stirred by roller shaker for 24 h at room temperature; and left at 4°C overnight to ensure a complete hydration prior to use in oscillation tests.

2.3. Small amplitude oscillatory shear measurements

Dynamic rheological measurements were conducted using HAAKE MARS III rheometer (Thermo Scientific, Karlsruhe, Germany) equipped with a Peltier plate for temperature control. A “parallel plate sensor” PP35/1Ti (diameter of 35 mm) was used for measurements.

Each sample was transferred onto the rheometer plate at the ambient temperature and excess material was wiped off with a spatula. In order to relax the samples before measurements, they were allowed to rest at the initial temperatures for 1 min. The RheoWin software 3.61 (Thermo Fisher Scientific) was employed for data evaluation. At least duplicate of each measurement were made.

Prior to making detailed dynamic measurements to examine the sample's microstructure, the linear viscoelastic region (LVR) must first be defined. The linear viscoelastic region (LVR) for PLSM samples was determined by performing an amplitude sweep measurements (0.01–100%) at constant frequency (1 Hz) and temperature (25 °C).

Frequency sweep tests at a constant strain in the LVE region were carried out to determine the viscoelastic nature of PLSM. In this test a strain of only 0.2% was applied in order to disturb the network formation the minimum possible. The mechanical spectra were characterized by G' and G'' [Pa] as functions of frequency in the range of 0.01 – 10 Hz at 25 °C. The storage and loss modulus account for the elastic and viscous component of a sample, respectively.

The temperature sweep measurements were performed at the constant strain of 0.2%, which was well within the linear viscoelastic region, while the frequency was fixed at 1.0 Hz.

The effect of temperature on rheological properties of PLSM was studied in four steps as follow; (a) linear increase in temperature from 20°C to 90°C (heating), (b) keeping at 90°C for 10 min, (c) linearly cooling down to 20°C (cooling) and (d) keeping at 20°C for 10 min. The heating and cooling steps were performed at three different heating rates of 1, 5 and 10°C/min,

2.4. Statistical analysis

Rheological properties of hydrocolloid solutions were determined by applying linear regression method based on minimizing sum of squares in Excel Microsoft Office software (v.2013). Model equations and regression coefficients (R^2) were reported and Analysis of variance (ANOVA) was applied to acknowledge any significant difference among rheological parameters; n and k at $P < 0.05$. The SPSS (version 13.0) program was used for all statistical analysis. All experiments were replicated minimum three times.

3. Results and discussion

3.1. Chemical composition

Chemical analysis indicated that the mucilage contained 4.01% protein, 3.17% moisture, 5.47% ash and no fat content. The values of ash and moisture contents were lower than the ones reported by Koocheki et al. [24] for *Eruca sativa* seed mucilage, Hosseini-Parvar et al. [25] for *Ocimum basilicum* L. seed mucilage and by Razavi et al. [26] for Basil seed gum. However, the protein and carbohydrate content observed was higher than *Ocimum basilicum* L. seed mucilage and Basil seed gum.

The total sugar content was 87.35%. PLSM samples comprise small amount of uronic acids (7.45%), revealing its weak polyelectrolyte nature and the relative amount of acidic polysaccharides in the mucilage. The uronic acid content of PLSM was higher than that of *A.homolocarpum* seed gum (5.63%) [1]. However, it was lower than that of gum Arabic (15.0%), xanthan gum (21.9%), flaxseed gum (21.0-25.1%), *C.pareira* pectin (70.56%), cress seed gum (15%), gum ghatti (12.83%), *A.bracteatum* gum (10.3%), sage seed gum (28.2-32.2%), and *Plantago major* seed mucilage (8.19%) [3,27–32].

3.2. Strain sweep measurements

In strain sweep measurements with raising strain, the two different domains namely linear viscoelastic and nonlinear regions were eminent. In the linear viscoelastic region, G' and G'' were nearly stable and in the nonlinear region, G' and G'' started to diminish. In the strain sweep measurements, G' remained constant until the strain reached a critical point at which G' began to decrease sharply (Fig.1). The strain at which G' decrease precipitously is called the critical strain. Therefore, critical strain reflects the deformability of the mucilage.

Strong gum solutions remain longer at linear state compared to weak gum solutions [33]; since the gum employed in this study is a strong gum, its viscoelastic moduli can be linear in a wide range of strain amplitudes. The linear region decrease respectively from gels to concentrated solutions and dilute solutions. While the strain value at the limit of LVR rarely exceeds 0.1% for colloidal gels, it may become equal to or above 1% for natural biopolymer gels [8].

The limiting values of strain (γ_L), $\tan \delta$ and τ obtained within the LVR ranges are tabulated in Table 1. At low mucilage concentrations (0.5-1%), the elastic modulus remained constant up to ca. 1% strain. With increasing mucilage concentration, the strain at which the elastic modulus decreased, increased to more than 1% (Table 1). This indicates that increasing mucilage concentration, would increase the strength of the system and make it more rigid. The limiting value of strain (γ_L) was high for 2% PLSM, implying a higher stability of the viscoelastic material under the γ -amplitude. Therefore, the strain of 0.2% for frequency sweep tests were well within the LVR, where the weak gel network was not damaged by the strain imposed during the measurements.

The values of G' and G'' at LVR also increased with increase in mucilage concentrations (Table 1). This type of test also determines the maximum deformation that a system can withstand before structural failure; in other words, determination of the critical strain within as corresponding of the LVR is a criterion of structural strength and shape retention versus the mechanical stresses. The stress corresponding to this strain is considered as yield stress (the tension that led to the first non-linear changes in the structure) [14].

Increasing mucilage concentration raise the yield stress values at flow point, meaning that the gel network became stronger (Table 1). The yield stress values for PLSM were 4.84, 5.53, 19.20 and 22.90 Pa for 0.5%, 1%, 1.5% and 2% mucilage concentrations, respectively. Yield stress has been considered as useful property of mucilage when they are used as binders, because it helps to cohere the different components of food formulations [34]. However, the magnitude of yield stress depends on the measuring technique and it is reported that yield stress values are actually defined by the rheological techniques and the assumptions made during the measurements [33,35]. The magnitudes of the viscoelastic moduli for PLSM were similar to previously reported values for some other food hydrocolloids such as *Alyssum homolocarpum* seed gum [1], *Lepidium perfoliatum* seed gum [14], maize starch pastes [36] and rice starch galactomannan mixtures [37].

3.3. Frequency sweep measurements

Frequency sweep information can be used to characterize or classify dispersions. The four most common and traditional classifications of dispersions are dilute solution, entanglement network systems (or a concentrated solution), weak gel and strong gel [8,33]. Fig. 2 shows

the changes in storage modulus (G') and loss modulus (G'') as a function of frequency (Hz) and concentration at 25°C. The PLSM had typical weak-gel like behavior where the magnitudes of G' and G'' slightly increased with raising frequency, having limited frequency dependency. According to Martínez-Ruvalcaba, Chornet, and Rodrigue [38], in weak gels, both moduli show slight frequency dependency, with G' exceeding G'' at all frequencies. The storage modulus (G') for PLSM was always higher than loss modulus (G'') within the experimental range of frequencies (0.01-10 Hz) and no crossover point occurred. Therefore, PLSM behaves more like a solid; that is, the deformations are elastic and recoverable. This behavior is compatible with that found in other hydrocolloids such as *psyllium* [39,40], rice starch-xanthan gum [41], basil seed gum [12], *Lepidium perfoliatum* seed gum [14] and *Alyssum homolocarpum* seed gum [1]. An increase in both storage and loss moduli were observed when concentration increased (Fig. 2), which is due to development of complex structures as concentration rises. The escalation in G' and G''

while being parallel, is connected with network defects [42]. In other words, at low concentrations, there are many intermolecular zones that do not participate in non-covalence cross junctions [42–44], while at high concentrations (1.5-2%), the maximum number of junction zones are formed. On the other hand, with increasing PLSM concentration, G' becomes greater than G'' indicating a clear tendency to form macromolecular networks. This type of behavior has also been reported by other authors for κ -carrageenan gels [45,46], gellan gum gels [47,48], κ -carrageenan/LBG gels [49,50], *psyllium* gels [40], *Lepidium perfoliatum* seed gum [14] and *Alyssum homolocarpum* seed gum [1].

Another characteristic value for evaluation of the viscoelastic behavior of mucilage is loss tangent ($\tan \delta$) which is the ratio of G''/G' . $\tan \delta$ is directly related to the energy lost per cycle divided by the energy stored per cycle. A $\tan \delta < 1$ indicates predominantly elastic behavior while $\tan \delta > 1$ indicates predominantly viscous behavior. In polymer systems, large $\tan \delta$ is an indication of dilute solutions while values between 0.2-0.3 reveal amorphous polymers and finally small $\tan \delta$ (near 0.01) suggest glassy crystalline polymers and gels [33]. Within the experimental frequency range (0.0628-62.8 rad.s⁻¹), the $\tan \delta$ values for samples of high concentration were between 0.18 and 0.25 which were much lower than those for low PLSM dispersions (Data not shown). This reveals that high concentration

samples may display an intermediate behavior between a weak and an elastic gel [51]. This result is similar to that obtained for other gums such as *psyllium* [40], *Lepidium perfoliatum* seed gum [14] and *Alyssum homolocarpum* seed gum [1].

PLSM showed a shear dependent flow behavior as the complex dynamic viscosity (η^*) decreased linearly with increasing frequency on a double logarithmic scale (Fig. 3). Therefore, we can conclude that mucilage dispersions had non-Newtonian shear-thinning behavior. Such behavior is consistent with those found for other dispersions [36,41,52–54]. The complex viscosity escalated as the mucilage concentration raised from 0.5 to 2% (Fig. 3). These results confirm the high potential of this hydrocolloid as a good thickener or stabilizer in increasing the consistency of food systems. Maskan and Gogus [55] also concluded that higher solid contents generally led to an increase in the viscosity due to the molecular movements and interfacial film formation.

From a structure point of view, for true gels, $\log G'$ versus $\log \omega$ plots have zero slope, while for weak gels and highly concentrated dispersions such plots have positive slopes and G' has a higher magnitude than G'' [8,56]. Based on polymer dynamics theory, for liquid-like fluids, the frequency dependency of G' values shows a power law relation [57]. Therefore, the power-law parameters used to model the frequency dependency of G' was evaluated using the following equation:

$$G' = a \cdot \omega^b$$

where G' is the storage modulus, ω is the oscillation frequency, and a is a constant. The exponent b is the slope in a log-log plot of G' versus ω . For a covalent gel, $b=0$ whereas for a physical gel $b > 0$. Low b values are characteristics of elastic gels, while for b values near 1, the system behaves as a viscous gel. At b values close to zero, G' does not change with frequency. The concentration significantly influenced the dynamic shear properties of the dispersions (Table 2). The frequency dependency of G' decreased when PLSM concentration increased. It is known that b value is related to the strength and nature of the gel [58]. This value also increased with increase in concentrations, which can be attributed to the formation of stronger network. Trckova, Stetina, and Kansky [59] also found that the elasticity of carrageenan-casein gels rose linearly with increasing carrageenan concentration.

3.4. Temperature sweep measurement

The temperature at which a biopolymer melts is defined as the temperature where the dynamic shear storage modulus (G') equals the dynamic shear loss modulus (G'') (the gel–sol transition)[60]. This can be easily measured using a controlled stress rheometer in oscillatory mode. It is important to recognize that viscoelastic moduli are frequency dependent and this dependence must be recognized when measuring the melting and gelling points [60].

The effect of temperature on G' and G'' of PLSM at different concentrations is shown in Fig. 4. The PLSM showed a storage modulus higher than the loss modulus, in the whole range of investigated temperatures. There was no crossover point for G' and G'' , which means the mucilage dispersion behavior was predominantly elastic and remains in the solid-like state.

During the initial heating (from 40 to 50 °C), G' decreased slowly as temperature increased, reaching a minimum around 50 °C (Fig. 4). The initial decrease in G' can be related to fluidity increase resulting from temperature raise. It may also be attributed to the energy dissipation from molecule movements and decrease in intermolecular interactions. This in turn reduce the energy required for flow, thus lowering the interference of the hydrodynamic domains [61,62].

The escalation of G' was observed to depend on PLSM concentration. With raising the temperature from 50 to 90 °C, the storage and loss moduli increased uniformly (Fig. 4). The G' increase might be due to formation of three-dimensional network structures and conversion of sol fraction into gel. In the PLSM dispersions, the G' increase while heating may also be caused by the thickening effect of the gums which restricts fluid mobility. Keeping the samples at 90 °C resulted in dramatic diminish of G'' and slight increase in G' , which suggests the presence of hydrophobic interactions. These interactions get robust by heating resulting in stronger interaction among PLSM polysaccharides molecules.

From rheological measurements of PLSM solutions, the hystereses between heating and cooling curves were found (Fig. 4). The storage modulus of PLSM decreased during cooling, which indicates that the hydrogen bonds were breaking and the network were getting weaker. However, Rafe and Razavi [12] and Hesarinejad et al [1] concluded that this increase in G'

during cooling was due to strengthened hydrophobic interactions and hydrogen bonds, respectively. Keeping temperature at 20°C had no significant effect on the storage and loss moduli. The hysteresis in the rheological measurements between the heating and cooling curves decreased by decreasing heating-cooling rates, indicating that the molecules of PLSM had enough time to form hydrogen bonds and to develop a firm and thermally irreversible network (Fig. 5).

4. Conclusion

The dynamic flow properties of PLSM within the linear viscoelastic region were investigated as function of mucilage concentration, temperature and heating-cooling rate. Aqueous dispersions of PLSM at different concentrations (0.5-2% w/v) showed similar viscoelastic behaviors. Both of the fluid-like (G'') and solid-like (G') behaviors were observed to be dependent on concentration and temperature. The elastic component of PLSM was consistently higher than its viscous component revealing its weak-gel like properties. It was further discovered/observed that the viscoelastic moduli of PLSM is only slightly dependent on frequency indicating that it is a cross-link network. From the rheological measurements, the hysteresis between the heating and cooling curves declined by decreasing heating and cooling rates, which shows that PLSM molecules had enough time to form hydrogen bonds and develop firm and thermally irreversible networks. The results revealed that PLSM had high total sugar content (87.35%), and it is likely an arabinoxylomannan-type polysaccharide. The monosaccharides composition demonstrated that PLSM is not a galactomannan or glucomannan. In conclusion, PLSM was stated to be an arabinoxylomannan-type polysaccharide with some compositional differences with many other seed gums. HPAEC analysis showed the presence of 69.42% Mannose as a main body with 11.98% and 7.42% Arabinose and Xylose, respectively as the main branches. Moreover, PLSM samples included a small portion of Glucuronic acids (7.45%), featuring its weak polyelectrolyte nature and the relatively moderate amount of acidic polysaccharides in the gum. These results argue the potential use of PLSM as a replacement for some of the plant hydrocolloids used in food industry at the moment.

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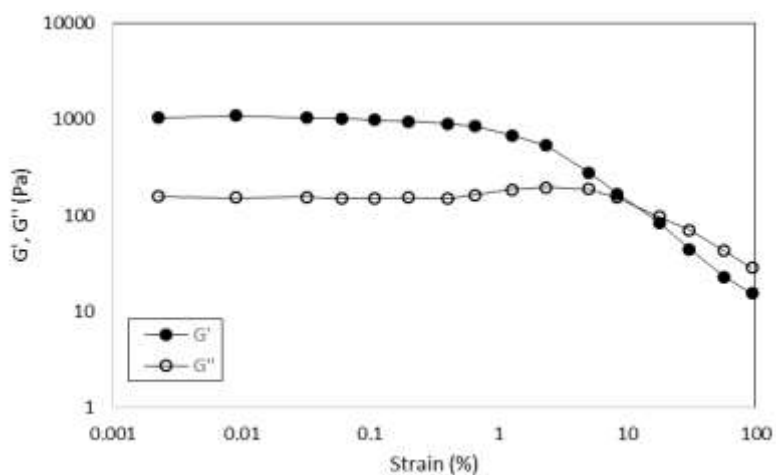


Fig. 1. Strain sweep dependency of storage modulus (G') and loss modulus (G'') for PLSM (2%) at 25 °C (frequency: 1 Hz).

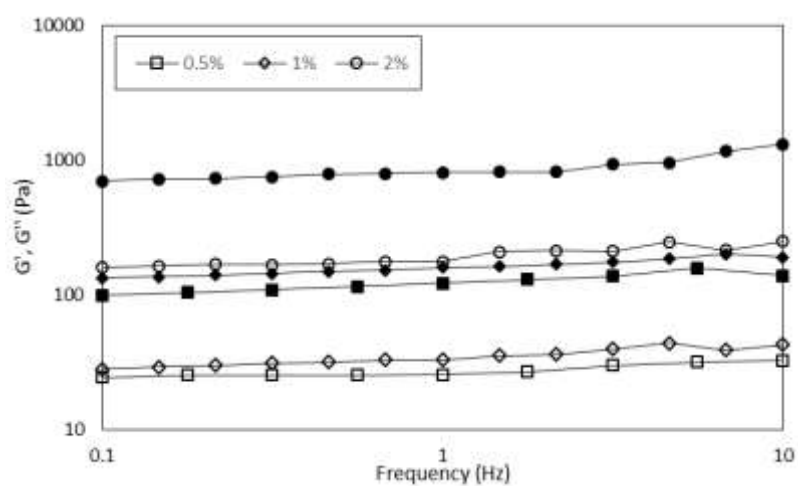


Fig.2. Effect of PLSM concentration on storage modulus (closed symbols) and loss modulus (open symbols) as functions of frequency at 25°C.

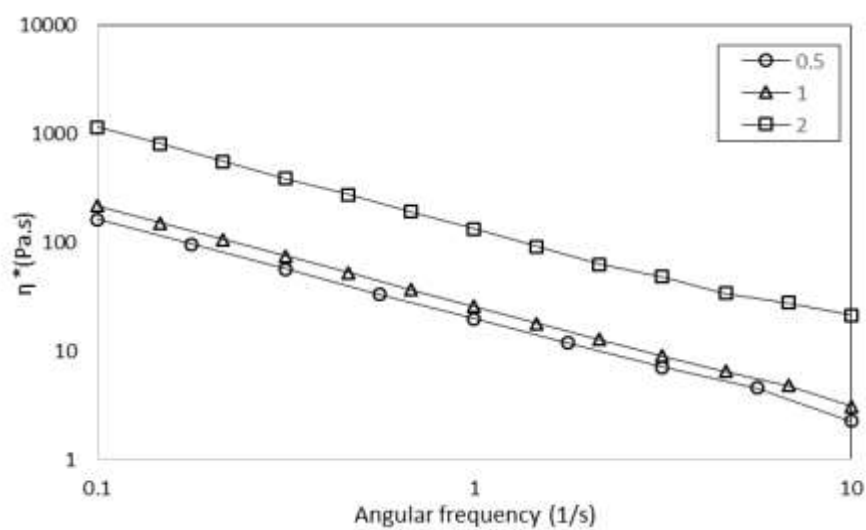


Fig.3. The effect of frequency on complex viscosity (η^*) of PLSM at different concentrations at 25 °C.

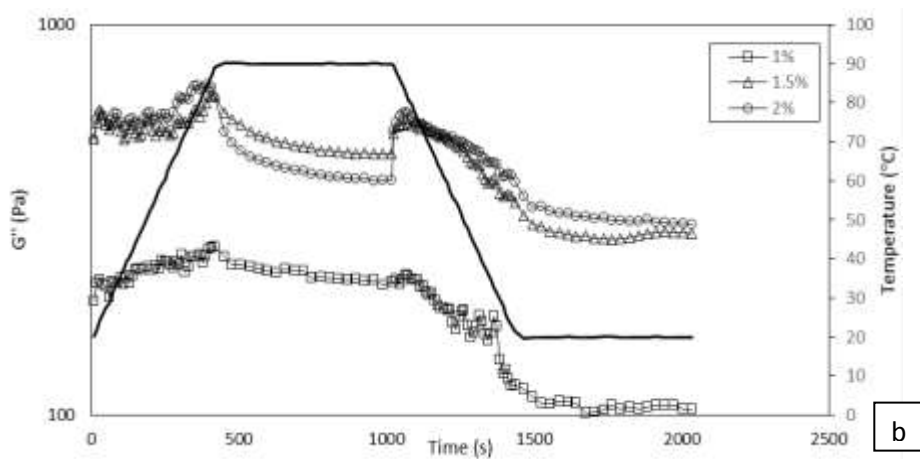
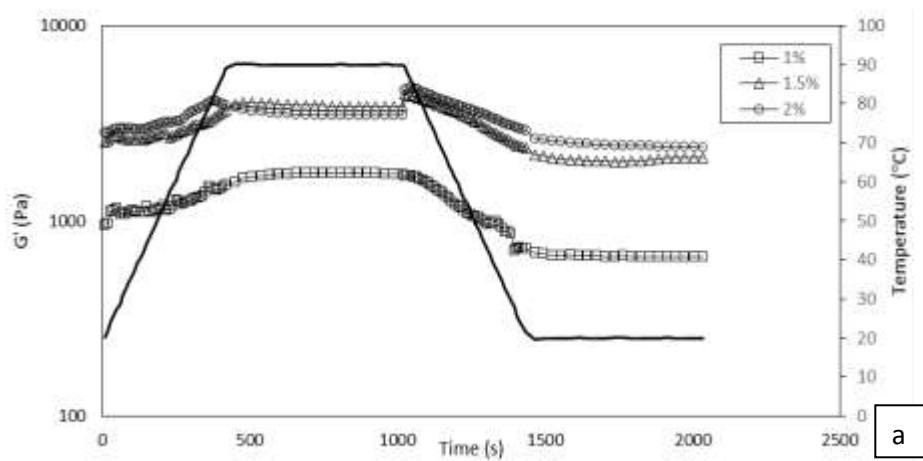


Fig. 4. Storage (a) and loss modulus (b) for different PLSM concentrations during heating cooling at the rate of 10 °C/min

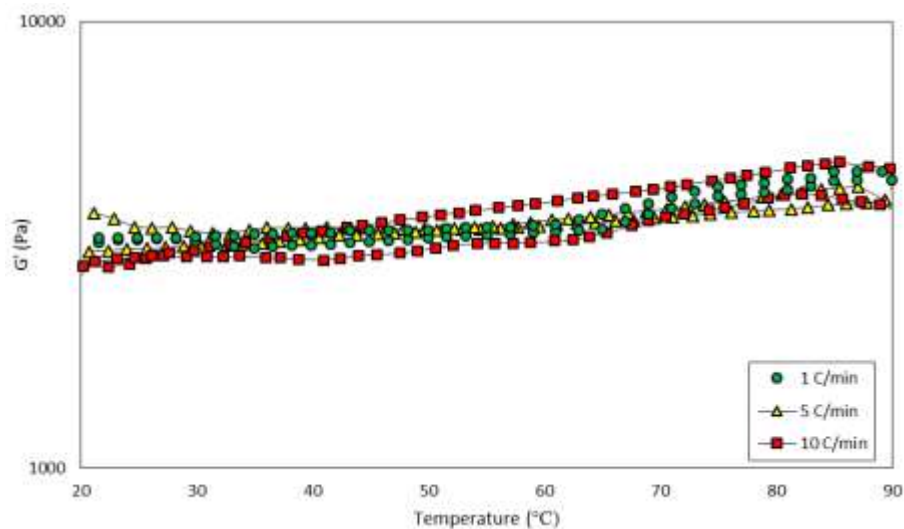


Fig. 5. Variation of storage modulus at different heating cooling rate of 1 °C/min, 5 °C/min and 10 °C/min at 2% PLSM concentrations.

Table1. Chemical composition of PLSM

Composition (%)*		
Carbohydrate		87.35±0.92
Monosaccharide		
	Arabinose	11.98±1.06
	Rhamnose	1.25±0.29
	Xylose	7.42±0.79
	Galactose	1.21±0.12
	Glucose	1.27±0.13
	Mannose	69.42±4.58
	Galacturonic acid	<MDL**
	Glucuronic acid	7.45±2.11
Protein		4.01±0.12
Moisture		3.17±0.21
Fat		<MDL
Ash		5.47±0.01

*Values are means ± SD of triplicate determination

**MDL: Method detection limits

Table 2. Structural strength G'_{LVE} , limiting value of strain γ_L , and loss-tangent value $\tan \delta_{LVE}$ in the linear viscoelastic range, yield stress at the limit of the LVE range, τ_y , and flow-point stress τ_f , with corresponding modulus $G_f : G' = G''$ for different PLSM concentrations at 25 °C, as determined by strain sweep tests (frequency: 1 Hz).

Mucilage Conc. (%)	G'_{LVE} (Pa)	G''_{LVE} (Pa)	γ_L (%)	Tan δ_{LVE}	τ_y (Pa)	τ_f (Pa)	G_f (Pa)
0.5	124.0	23.8	0.73	0.19	0.92	4.84	19.4
1	157.4	32.3	1.04	0.20	1.18	5.53	21.2
1.5	419.2	79.3	3.80	0.19	3.69	19.20	48.9
2	842.5	164.2	6.61	0.19	5.67	22.90	90.4

Table3. Power-law parameters for the storage modulus at 25 °C.

Mucilage Conc. (%)	b	a	R^2
0.5	0.10	122.7	0.90
1	0.09	160.5	0.97
1.5	0.09	429.5	0.83
2	0.08	859.1	0.91