Sensory texture based on rheological properties: Applications in model foods and cheese

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ABSTRACT

A series of gels were used as model foods to understand relationships among rheological properties and sensory texture. Fracture stress was the best predictor of sensory fracture force and particle breakdown, whereas a measure of the non-linear region was a better predictor of sensory deformation.

INTRODUCTION

Food texture is a complex property involving a constant evaluation of the rheological and tactile properties during oral processing. To understand the role of rheological properties in food texture, we developed a series of four model food gels (polyacrylamide, alginate, agarose and agar) that allowed for precise manipulation and measurement of mechanical properties. All four gels can be made from aqueous solutions with polymer concentrations similar to what is found in foods. Polyacrylamide gels were used as a starting point because they show linear elasticity that is independent of strain level and rate, and can be modeled based on rubber elasticity. With that in mind, gel firmness/hardness could be increased by addition of more polymer or increased crosslinking. The three other gels were made with polysaccharides and all showed non-linear viscoelastic behavior. Alginate gels were investigated because they are food-grade and, because calcium interacting with uronic acids of the polymer chains causes cross-links,[1] the addition of different amounts of calcium provides a way to compare with the covalently cross-linked polyacrylamide gel. Agarose and agar gels are formed by non-covalent association among strands (physical gels) and thereby contrast with the covalently linked polyacrylamide gels (chemical gels). Agar gels are also food grade and were evaluated by a trained sensory panel in addition to mechanical analysis.

RHEOLOGICAL CHARACTERIZATION

To investigate important mechanical properties of food gels and the impact they likely contribute to overall product texture, a few critical conditions must be considered, namely extent and speed of compression and shear. Ignoring the breakdown contributions from thermal and enzymatic factors, oral processing is mainly dominated by a chewing action. This process of destruction can occur at varying rates, but ultimately a large gel is reduced to many smaller pieces that are conducive with swallowing. Rheologically, mastication is a complex set of actions involving shear and compressive stresses. Each of these stresses may occur at varying speeds. As the stresses are applied to the sample, it undergoes a strain. Understanding the stress and strain relationship permits computation of important mechanical properties, like viscoelastic moduli and fracture properties.

To begin to fundamentally describe the mastication process, instrumentation is used to subject the gel to either a shear or compressive type of load. With respect to the condition of shear, rheometry permits the measurement of viscoelastic moduli (G'

and G") by gently probing within the linear viscoelastic regime. However, small amplitude oscillatory shear testing seldom correlates with sensory texture[2]. To extend rheological shear measurements into the realm of non-linearity and fracture, samples must be compressed, extended or twisted to the point of fracture. A method that fits well with highly deformable gels (valid for strains up to 3.0) is torsion analysis. Torsion deforms samples with a twisting action at controlled rates to a condition of macroshear, allowing for determination of the non-linear and failure characteristics. Equations for torsion stress and strain are based on calculations described by Diehl et al.[3] and Truong and Daubert[4]. The non-linear viscoelastic region was mathematically modeled using two equations. The first model is a strain energy density function proposed by Blatz et al.[5], commonly referred to as the BST equation. This model is based upon a relationship between deformation energy and non-linear elasticity. For torsion conditions of pure shear, the shear stress (σ) is predicted by

$$\sigma = \frac{2G_{BST}}{n}\left(\lambda^n - \lambda^{-n}\right) \qquad (1)$$

and,

$$\lambda = \frac{\gamma + \sqrt{\gamma^2 + 4}}{2} \qquad (2)$$

where G_{BST} and n are fitting parameters and λ is a stretch ratio based on shear strain (γ). An alternative, second order polynomial equation was also considered to incorporate the linear and non-linear rheological behavior

$$\sigma = G_{poly}\gamma + k\gamma^2 \qquad (3)$$

where k represents the rate of slope change from the non-linear regime

$$k = \frac{G_A - G_{poly}}{\gamma_A} \qquad (4)$$

and G_A is the shear modulus at a corresponding shear strain (γ_A). G_{poly} is a constant representing the shear modulus from the initial stress-strain curve.[6]

RHEOLOGICAL PROPERTIES OF GELS

Polyacrylamide gels

These gels are linearly elastic up to fracture and not strain rate dependent (Fig. 1)[7,8]. In gels with a fixed monomer to cross-linker ratio (40:1), fracture stress increases with increasing polymer concentration, whereas fracture strain remains constant.[8]
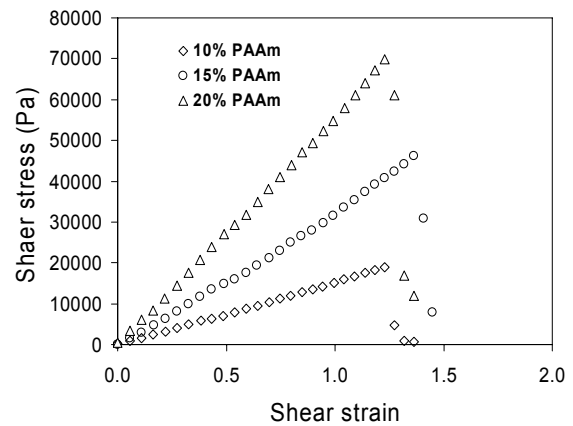

Figure 1. Linear fracture behavior of polyacrylamide gels made at different polyacrylamide (PAAm) concentrations. Reprinted from reference 8 (Figure 11) with kind permission of Springer Science + Business Media. © Springer-Verlag 2005.

Fracture of acrylamide gels demonstrated two properties that are consistent with other food gels. The first being an increase in network concentration causes an increase in fracture stress. The second observation is that fracture strain is determined more by the properties of the network structure than the amount of network structure.

Glycerol and dextran were added to examine the effects of solutes that can cause an increased in viscosity of the fluid phase surrounding the gel network. Neither solute causes a significant change in the small strain rheological properties (G'); however, there is an increase in fracture stress and strain.[9] This shows that glycerol and dextran do not alter the basic nature of the gel network (elastic properties), but do change the fracture process. The increase in fracture stress and strain are associated with

a switch in fracture mechanisms from brittle to elasto-plastic.[9] This transition decreases the stress concentration factor at the tip of the propagating fracture, thereby increasing the amount of stress required to fracture the material.

Alginate gels

Acrylamide gels were used to show the effects of changing network composition in a linear elastic material. In contrast, alginate gels are viscoelastic and at large deformations show non-linear viscoelastic behavior, similar to most food gels. At a fixed calcium:alginate ratio, increasing alginate concentration causes an increase in fracture stress while fracture strain remains relatively constant; similar to what was observed for acrylamide gels (Fig. 2).[8] When alginate concentration is constant and calcium concentration increases, fracture strain remains essentially constant and at the same value as when the overall concentration of alginate was increasing (Fig. 2). Fracture stress increasing with calcium concentration is similar to what is observed for increasing alginate concentration at a fixed calcium:alginate ratio, with a deviation at higher fracture stresses.
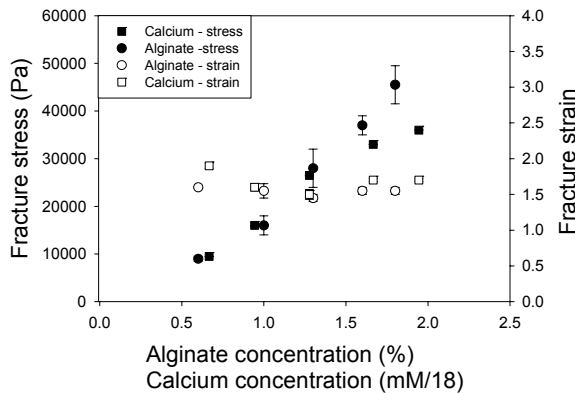

Figure 2. Fracture stress and strain of alginate gels when different amounts of crosslinker (calcium) are added, or when calcium:alginate ratio (0.038 w/w) is constant and overall polymer concentration increased. Data from reference 10.

Fracture stress of alginate gels initially increases then plateaus as strain rate is

increased.[10] Fracture strain is also strain rate dependent but to a lesser degree. Unlike acrylamide gels, alginate gels show strain hardening before fracture. The degree of strain hardening can be modeled using the BST equation as described in the *rheological characterization* section. As seen in Fig. 2 and Table 1, the BST constant and fracture strain remain relatively unchanged as fracture stress increases 4 to 5 fold. Moreover, there appears to be no major difference in strain hardening if the gel network or crosslinker concentrations are increased. The data in Fig. 2 and Table 1 suggest that gels with fracture stresses ranging from ~10 to 40 kPa, fracture strain of ~1.6 and a BST constant around 5.5 can be made by either adjusting the amount of calcium (fixed polymer) or polymer (fixed calcium:polymer ratio). While these gels are very similar in large strain rheological properties, it remains to be seen if they would have the same sensory texture.

Table 1. Changes in BST constant (n) and fracture strain (γ_f) with an increase in calcium at constant polymer concentration or an increase in alginate concentration at constant calcium:alginate ratio (0.038 w/w). Data from reference 10.

Calcium (mM)	n	γ_f		Alginate (%)	n	γ_f
11.8	4.6	1.96		0.5	5.4	1.66
17.6	5.2	1.62		0.75	5.3	1.60
23.5	5.4	1.51		1.00	5.6	1.50
29.4	5.4	1.63		1.25	5.6	1.57
35.3	5.2	1.64		1.5	5.6	1.59

Agarose gels

These gels were used as a model for a non-linear viscoelastic physical gel (non-covalently linked network). Like polyacrylamide and alginate gels, increasing network concentration increases fracture stress (Fig. 3). Unlike polyacrlamide and agarose gels, fracture strain decreases as concentration increases. Addition of glycerol increases linear viscoelastic properties (G', data not shown), fracture stress (Fig. 3) and fracture strain (with the

exception of the lowest strain rate; Fig. 4)[6]. In this case, glycerol is altering the basic gel network and the fracture properties, as compared to acrylamide gels where just the fracture properties were changed. The fracture properties of agarose gels also depend on strain rate. As strain rate increases, the fracture stress has a major increase, while fracture strain increases slightly; similar to what was observed in alginate gels (Fig. 3 and 4).[10] Like acrylamide and alginate gels, the fracture strain was less affected by network concentration than fracture stress. The non-linear behavior of agarose gels is described better by the polynomial than the BST model; and the fitted parameter, k, is strain rate dependent.[6]
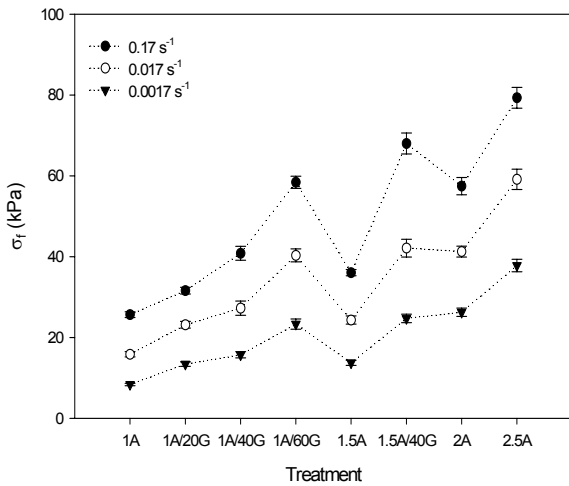

Figure 3. Fracture stress (σ_f) of agarose gels containing different levels (w/w) of agarose (A) or glycerol (G). Symbols indicating different strain rates are shown in insert. From reference 6 with permission from Elsevier.

Gel rheological fingerprint

The above discussion described how rheological and fracture properties change among a series of gels. Information can be obtained at three different strain levels outlined in Fig. 5. The linear region provides information on the network structure that can be expressed as G', G" and phase angle (δ).

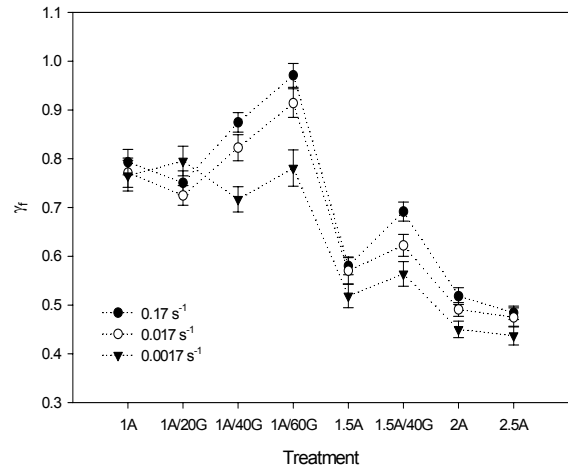Figure 4. Fracture strain (γ_f) of agarose gels containing different levels (w/w) of agarose (A) or glycerol (G). Symbols indicating different strain rates are shown in insert. From reference 6 with permission from Elsevier.

Once strain exceeds the linear viscoelastic region but is lower than fracture strain, there are various models that can be used to characterize the non-linear region (e.g, n_{bst} and k_{poly}). Finally, the fracture stress (σ_f) and strain (γ_f) can be measured.
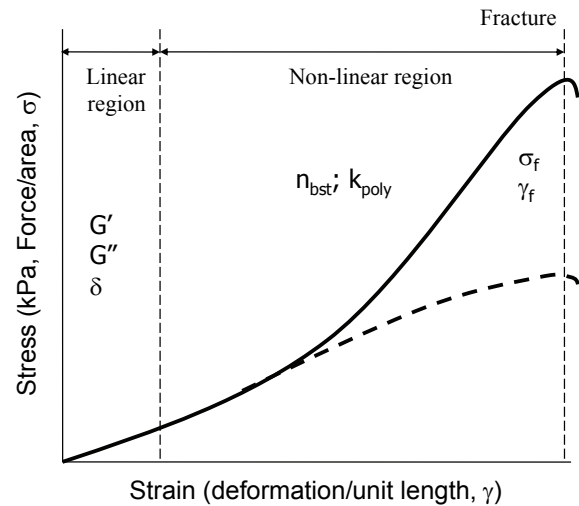

Figure 5. Hypothetical stress-strain relationship for a gel at a constant strain rate showing strain hardening (solid line) or weakening (dashed line).

A full characterization of a viscoelastic material requires determining strain rate effects. Changes in fracture properties as a function of strain rate can be used to

characterize a viscoelastic gel based on the energy model proposed by van Vliet et al.[11]

$$W = W' + W''_m + W''_c + W_f \qquad (5)$$

This model stipulates that the energy (W) supplied during deformation is either stored (W') or dissipated by various processes, or used to break network structures. Energy can be dissipated due to friction between components arising from inhomogeneous deformation (W''_c), by viscous flow (W''_m) or used to cause fracture (W_f).[11] An increase in strain rate is predicted to cause an increase in fracture stress for both W''_m and W''_c mechanisms. In contrast, fracture strain should decrease for W''_m and increase for W''_c. This equation shows that the molecules composing the gel network, the structure of the gel network, and interactions between molecules have various ways to influence the fracture process.

Some general conclusions can be made about mechanical properties of acrylamide, alginate, agarose and agar gels that are relevant to food texture. Fracture stress can be increased by increasing the amount of network (i.e., network density) or the degree of crosslinking within the network. When deformed, alginate, agarose and agar gels all have energy being dissipated due to friction between components arising from inhomogeneous deformation (W''_c), causing fracture stress and strain to increase with strain rate.[6, 10, 12] Lastly, non-linear behavior can be accurately modeled; however, neither of the two models tested are universal enough to fit the two gels with equal accuracy. A recent study[13] compared 11 models (including BST) for stress-strain behavior of gellan gels and concluded that only three empirical equations adequately describe the data. Therefore, a "rheological fingerprint" of these gels would include the following: linear viscoelastic properties, a measure of the non-linearity from the appropriate model, and fracture stress and strain, all as a function of strain rate.

RELATING RHEOLOGICAL FINGERPRINTS TO SENSORY TEXTURE

Descriptive sensory analysis is a technique where panelists are trained to detect specific textural properties. For example, in evaluating the textural property of *hand fracture force*, the panelists are instructed to "press the sample between the thumb and first two fingers until sample fractures."[2] The panelists score the amount of force as 1 to 10 relative to a range of standard materials. Descriptive analysis was used to quantify hand texture characteristics of agarose gels, and correlations with σ_f (Fig. 3), γ_f (Fig. 4), fracture modulus (G_f, fracture stress/fracture strain, Fig. 6), G', G" and G* are shown in Table 2. Linear viscoelastic moduli and fracture properties were determined at relative strain rates of $0.31\ \text{s}^{-1}$ and $0.17\ \text{s}^{-1}$.

Table 2. Correlation coefficients (r-values) among sensory and rheological terms for agarose gels. Highest correlations are underlined. *Denotes $p \leq 0.05$. **Denotes $p \leq 0.001$. Data from reference 2.

Rheological Property	Sensory Term		
	Small-Strain Force	*Fracture Force*	*Fracture Deformation*
G'	0.68	0.75	-0.23
G"	0.60	0.67	0.04
G*	0.61	0.69	-0.15
σ_f	0.76*	0.82*	-0.16
γ_f	-0.68	-0.61	0.98**
G_f	0.98**	0.99**	-0.67

Four texture attributes were used to describe the gels, including *hand small-strain force, hand springiness, hand fracture force,* and *hand fracture deformation*. All gels had the same level of springiness. Small-strain force and fracture force terms were capable of differentiating each of the gel treatments seen in Fig. 3 and 4.[2] Surprisingly, the hand force terms correlate more highly with fracture modulus values than fracture stress values (Table 2), suggesting hand perception of force includes a coupling of stress and strain. The changes in G_f across the treatments are seen in Fig. 6. This property

shows a steady increase as shear rate increases. It should be noted that correlations can be coincidental or reflective of cause and effect. Fig. 7 shows that increasing agarose concentration increases G_f and decreases γ_f, while increasing glycerol concentration increased G_f and γ_f, meaning there was a series of gels with similar G_f (~30 to 60 kPa) and a range of γ_f (~0.58 − 0.98). This mixture of fracture properties in gels was designed to diminish the possibility of coincidental correlation.
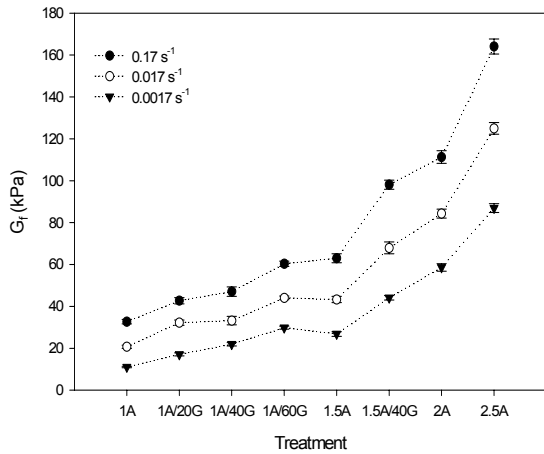

Figure 6. Fracture modulus (G_f) of agarose gels containing different levels (w/w) of agarose (A) or glycerol (G). Symbols indicating different strain rates are shown in insert. From reference 6 with permission from Elsevier.

Hand fracture deformation was correlated with γ_f and no other mechanical property. Rheological properties determined in the linear viscoelastic region (G', G" and G*) are not significantly correlated with any texture term, including small-strain force. This response demonstrates that even relatively small deformations between fingers (for this term, the panelists were instructed to "press sample between thumb and first two fingers until it is depressed 10% of original height") are not detecting what is occurring in the linear viscoelastic region.

Since agarose gels are not food-grade and therefore could not be ingested, an additional investigation using agar gels was undertaken. Descriptive sensory analysis of agar gels was done using a broader range of

texture terms that encompassed the mastication process. The first three terms (*small-strain force, fracture force* and *deformability*) were similar to the hand terms used with agarose gels but were evaluated in the mouth by pressing between the molars.[12] The *particle breakdown* term evaluated the number of particles after three chews and the *chewiness* term determined the number of chews required to prepare the sample for swallowing. These terms are associated with the physiological concept of oral processing, which is the process used to prepare food for swallowing[14].
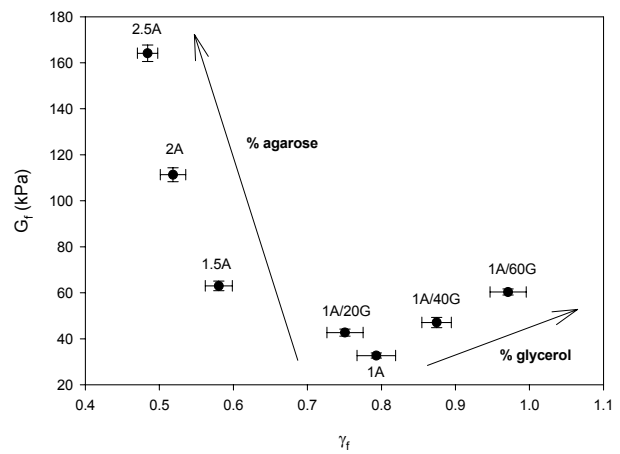

Figure 7. Fracture modulus (G_f) of agarose gels containing different levels (w/w) of agarose (A) or glycerol (G). From reference 2 with permission from Elsevier

Table 3. Correlation coefficients (r-values) among sensory and rheological terms for agar gels. Abbreviations are: *ssf*, small-strain force; *ff*, fracture force; *def*, deformability; *pb*, particle breakdown and *chew*, chewiness. Highest correlations are underlined. *Denotes p ≤ 0.05. Data from reference 12.

Rheol. Property	Sensory Term				
	ssf	*ff*	*def*	*pb*	*chew*
σ_f	<u>0.95*</u>	<u>0.96*</u>	-0.59	<u>-0.97*</u>	<u>0.97*</u>
γ_f	-0.38	-0.37	0.88*	0.23	-0.35
G_f	0.93*	0.94*	-0.87*	-0.86*	0.95*
G_{poly}	0.90*	0.91*	-0.87*	-0.83*	0.94*
k	0.85*	0.84*	<u>-0.93*</u>	-0.74	0.84*

All sensory terms evaluated are significantly correlated (positively or negatively) with a mechanical property at r

$\geq$ 0.93 (Table 3). Similar to the hand evaluation of agarose gels, small-strain force and fracture force of chewed agar gels are significantly correlated with G_f and σ_f. However, the level of correlation is reversed, with closer correlations between σ_f and force-related sensory properties than G_f (Table 3). The reason for this difference is not clear and may be explained by differences in physiological detection of force-deformation relationships.

The deformability of agar gels is not surprisingly correlated with γ_f and terms containing strain (G_f, G_{poly}), but also with a measure of the non-linear region (*k*). This suggests that gels with a high degree of strain hardening are viewed as being less deformable. The extent of particle breakdown is correlated negatively with terms indicating a stronger network (σ_f, G_f, and G_{poly}), and the number of chews were positively correlated with the same mechanical properties in addition to the non-linear behavior. Taken as a whole, the sensory texture of agar gels can be accurately described by fracture stress and a non-linear constant representing the degree of strain hardening (Table 3).

Cheese texture

From the previous discussion it is clear that the sensory texture properties of agar gels are easily predicted based on rheological properties. Can the same be said for cheeses with similar fracture properties? Changes in sensory texture and rheological properties of Monterey Jack and Mozzarella cheese were measured over 38 days of storage[15]. For both cheeses, σ_f ranged from 22 to 32 kPa and γ_f ranged from 1.1 to 2.0 (determined at a strain rate of 0.47 s^{-1})[15]. As can be seen in Table 4, the general trend is for fracture properties to be more predictable of sensory texture than linear viscoelastic properties. What is not shown in the table are three textural properties evaluated during chewing (cohesiveness, smoothness and smoothness of mouth coating) that were not correlated with either linear viscoelastic or fracture properties. These textural elements are not found in agar gels but are key to cheese texture. Moreover, they tend to be associated with particle breakdown and particle characteristics that may be difficult to detect by rheological methods.

Table 4. Correlation coefficients (r-values) among sensory and rheological terms for Monterey Jack and Mozzarella cheese. Abbreviations are: *hff*, hand fracture force; *hsp*, hand springiness; *ff*, fracture force; *pb*, particle breakdown and *adh*, particle adhesiveness to mouth surface. *Denotes $p \leq 0.05$. **Denotes $p \leq 0.005$. Data from reference 15.

Rheol. Property	Sensory Term				
	hff	*hsp*	*ff*	*pb*	*adh*
G'	0.62*	-0.30	0.50*	0.20	0.34
G"	0.38	0.07	0.40	0.03	0.11
G*	0.62*	-0.30	0.50*	0.20	0.34
δ	-0.82**	0.47	-0.67	-0.14	-0.31
G_f	0.66**	-0.83**	0.32	0.48*	0.65**

SUMMARY

Model gels prepared from acrylamide, alginate, agarose and agar provide a comprehensive way to understand molecular mechanisms responsible for gel texture. Factors determining fracture stress and strain are established for all gels. Mathematical models for non-linear behavior are useful in describing the degree of strain hardening. For agar gels, a combination of fracture properties and non-linear behavior accurately describes sensory texture. In contrast, cheese is a more complex material that required further investigation, especially concerning the breakdown during chewing.

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