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## Characterization of polyacrylamide gels as an elastic model for food gels

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**Abstract** Serving as an elastic model system for food gels, characteristics of polyacrylamide (PAAm) gels were investigated using small amplitude and large deformation rheological tests. The PAAm gels displayed elastic or viscoelastic behavior depending on network crosslink density. For elastic PAAm gels, the rheological properties obeyed the theory of rubber elasticity; whereas for viscoelastic PAAm gels, shear modulus depended on temperature. The elastic PAAm gel fracture parameters did not change with deformation rate ( $0.2\text{--}5.5\text{ s}^{-1}$ ), indicating insignificant viscous flow during deformation. Fracture stress was correlated with gel monomer concentration, whereas the fracture strain remained constant regardless of the monomer concentration. In

addition, the stress was linearly proportioned with strain up to fracture, indicating that PAAm gels did not experience finite network chain extensibility during large deformation. Consequently, the fracture of PAAm gels did not result from the extensional limitation of network chains, nor did gel fracture result from the nonlinear force–distance relationship between polymer connections. Purportedly, the fracture of PAAm gels was caused by external force overcoming the gel cohesive forces, and low strength of PAAm gels compared to rubbers caused fracture prior to experiencing nonlinear stress-strain deformation.

**Keywords** Polyacrylamide gels · Food gels · Fracture property · Large deformation

### Introduction

Many popular foods such as gelatin desserts, frankfurters, surimi-based shellfish analogs, and cooked egg white are gels or gel-based foods. A common characteristic among these foods is that they are primarily composed of liquids but respond as solids, displaying a high degree of elasticity. When continuously deformed, these materials will eventually fracture. Fracture is inherent throughout food processing applications, from manufacturing to mastication. Consequently, researchers frequently attempt to relate the fracture behavior of gels to sensory texture (Wium et al. 1997; Drake et al. 1999; Antoniou et al. 2000; Truong et al. 2002; Foegeding et al. 2003).

Certain sensory texture notes have been significantly correlated with fracture properties (Montejano et al. 1983; DeMars and Ziegler 2001; Lillford 2001; Brown et al. 2003; Foegeding et al. 2003), supporting the concept that a fundamental understanding of food texture can be obtained by investigating mechanisms responsible for fracture behavior (Foegeding et al. 1994; van Vliet and Walstra 1995).

Food gels are complex and limited in determining mechanisms responsible for large deformation and fracture properties. One reason is that food gels are viscoelastic, and part of the energy from the applied force dissipates as heat and the fracture behavior fluctuates with deformation rate, complicating interpretation of the

fracture mechanisms (Foegeding et al. 1995). Another reason is that food gels are compositionally complex, hampering a detailed analysis of the structural elements affecting fracture. For instance, the crosslinks in food gels can be chemical and/or physical, and the network structure of the gels is quite complicated and difficult to control and quantify. The less-defined networks make any theoretical description of rheological properties for food gels quite challenging. Therefore, fracture properties will be best understood using simple materials with well-defined chemical and physical properties (Foegeding et al. 1995), and the simplest gel models can be derived from simple polymers with permanent crosslinks at random positions (Edwards et al. 1987).

Polyacrylamide (PAAm) gels were proposed as elastic model systems to study gel behavior during large deformation (Foegeding et al. 1994). PAAm gels are formed by a small amount of matrix material stabilizing a large amount of water, similar to food gels. PAAm gels are covalently cross-linked materials, which are well understood on a chemical and physical basis, and PAAm gels have been used as models in many physical investigations (Nossal 1985). As elastic gels, the widely acknowledged theory of rubber elasticity could be used to characterize gel structure based on rheological measurements.

The network formation of PAAm gels depends on many factors such as the monomer (acrylamide) and crosslinker (*N,N'*-methylene-bis-acrylamide) concentration (Flory 1953; Odian 1970). Mechanical measurements of PAAm gels have focused on the small deformation region using various techniques (Richards et al. 1977; Janas et al. 1980; Nossal 1985), and the shear modulus of PAAm gels depends on the crosslink density and the number of equivalent primary chains (Nossal 1985).

Having elastic properties, some engineering polymers (natural rubbers and synthetic polymers) have been studied extensively (Bot et al. 1996). The fracture of engineering polymers normally occurs following strain-hardening behavior (Treloar 1975). Strain hardening means the stress-increasing rate increases with deformation magnitude. Therefore, to understand the reason for strain hardening may help to explain the mechanisms for fracture. In general, material failure occurs when bonds between the structural elements break, resulting in a breakdown of the structure over the length scales much larger than the structural elements (van Vliet and Walstra 1995). From a molecular point of view, the explanation for this strain-hardening behavior was based on the postulate of finite extensibility of polymer chains, achieved at large deformation (James and Guth 1943; Kilian 1981; Edwards and Vilgis 1986; Kluppel 1994; Spathis 1995). Finite chain extensibility means that when the strain in a network exceeds a fraction of the maximum deformability of the network, a nonlinear stress-strain response will result. Consequently, the

Gaussian theory becomes inadequate in predicting deformation (Treloar 1975). Bueche and Halpin (1964) suggested that network chains of gum elastomers first become fully extended (after viscoelastic rearrangement), and then break. The theory of Bueche and Halpin allows the possibility of relating large deformation behavior and fracture properties to network chain length and bond strength. Groot et al. (1996) developed a mesoscopic model for the endpoint separation dependent scission rate of a polymer connection in a network, and the theory was applied to gelatin. The results showed that the nonlinear force-distance relationship of the polymer connections between crosslinks was a reason for the early and sudden fracture of polymer gels.

The association of fracture properties of PAAm gels with temperature and relaxation characteristics were previously investigated (Foegeding et al. 1994). Other aspects, particularly how the rheological and fracture properties relate to gel network crosslink density and network chain length, have not been reported. The relationship between fracture properties and gel networks in PAAm may help to explain fracture mechanisms in food gel systems. Therefore, the gel fracture properties associated with gel networks were investigated in this paper.

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## Materials and methods

### Gelation of PAAm gels

Gels were prepared by co-polymerization of acrylamide (Sigma Chemical Co., St. Louis, MO) as the monomer with *N,N'*-methylene-bis-acrylamide (Sigma Chemical Co., St. Louis, MO) as the crosslinker in aqueous solution. The ammonium persulfate/*N,N,N,N'*-tetramethyl ethylenediamine (TEMED) (Bio-Rad, Hercules, CA) redox system was used to initiate polymerization at room temperature. The gels containing a given monomer concentration but differing amount of crosslinker, or a given monomer to crosslinker (*M/C*) ratio but differing monomer concentration, were prepared from stock solutions of acrylamide (30% w/v) and bis-acrylamide (3% w/v). The acrylamide stock solution was prepared by dissolving acrylamide crystal granules in distilled water under constant stirring. The stored stock solution was kept in opaque plastic bottles to prevent decomposition by light. The stock solution of bis-acrylamide was made using the same procedures as acrylamide. Next, the solutions of varying monomer or crosslinker concentration were prepared by mixing appropriate amounts of the two stock solutions with water to adjust the solutions to the final volumes. Ammonium persulfate (0.075% w/v in the final solutions) was added, followed by addition of TEMED (5 ppm v/v in the final solutions).

### Small strain tests

The gels were made in situ for small amplitude rheological tests at room temperature. The aforementioned gel solutions were quickly poured into the concentric cylinder cup (CP 25) of a Bohlin VOR rheometer (Bohlin Rheologi AB, Lund, Sweden) to gel for at least 8 h at room temperature. The couette system with serrated surfaces was used to minimize slip of materials on sidewalls. Rheological properties were measured within the linear viscoelastic range using an oscillatory test with a strain amplitude of 0.0206 and a frequency of 0.05 Hz. Strain sweeps were used to measure the small strain shear modulus change with strain at different frequencies (from 0.01 Hz to 20 Hz). Unless stated otherwise, all tests were performed at  $25 \pm 0.1^\circ\text{C}$ .

The number of elastically active network chains (EANC) per unit volume ( $N$ ) for rubber-like (entropy-based) materials were calculated according to the theory of rubber elasticity (Treloar 1975),

$$G = gRTN, \quad (1)$$

where  $G$  is the equilibrium small strain shear modulus;  $g$  is a constant, which is close to 1 for incompressible materials;  $R$  is the gas constant (8.314 J/mol.K);  $T$  is the absolute temperature (K). The network chain molecular weight, corresponding to the network chain length, was calculated by  $N = C/M_c$ , and expressed as (Treloar 1975; Oakenfull 1984),

$$G = \frac{CRT}{M_c}, \quad (2)$$

where  $C$  (g/m<sup>3</sup>) is the sample concentration, and  $M_c$  (g/mole) is the average molecular weight of each chain segment in the network.

For creep recovery experiments, a 5% PAAm gel ( $M/C$  37.5) was made in the same procedure as mentioned for the Bohlin Rheometer but in a serrated couette system attached to a Stresstech rheometer (Rheological Instruments AB, Lund, Sweden). A 200 Pa stress was applied to the gel for 100 s, then removed. The strain change with time was recorded. The test was at  $25 \pm 0.1^\circ\text{C}$ .

For determination of gel linear viscoelastic regions and testing gel rheological properties (elastic or viscoelastic), gels were made as previously described in a serrated couette system attached to a Bohlin VOR Rheometer (Bohlin Rheologi AB, Lund, Sweden). For determination of gel linear viscoelastic regions, shear storage modulus  $G'$  and shear loss modulus  $G''$  associated with shear strain at a frequency of 0.1 Hz was measured. For testing the gel rheological properties,  $G'$  and  $G''$  associated with frequency were measured. A constant strain magnitude of 0.02, which was in the gel linear viscoelastic region, was used. If the shear storage

modulus  $G'$  was frequency dependent, the gel was deemed viscoelastic. Otherwise, the gel was considered elastic. All measurements were performed at  $25 \pm 0.1^\circ\text{C}$ .

### Torsional fracture tests

Capstan-shaped PAAm gels were prepared as described by Foegeding et al. (1994). Gels were fixed onto notched styrene disposable disks with cyanoacrylate glue (Quik Tite, Loc Tite, Rocky Hill, CT) and mounted onto a Haake VT550 viscometer (Paramus, NJ) for twisting at a specified rotational speed. True shear stress and strain at fracture were calculated from the respective torque and angular displacement data, using a modified version of the calculations described by Diehl et al. (1979) and Truong and Daubert (2000). Torsion analysis was performed for five samples of each treatment.

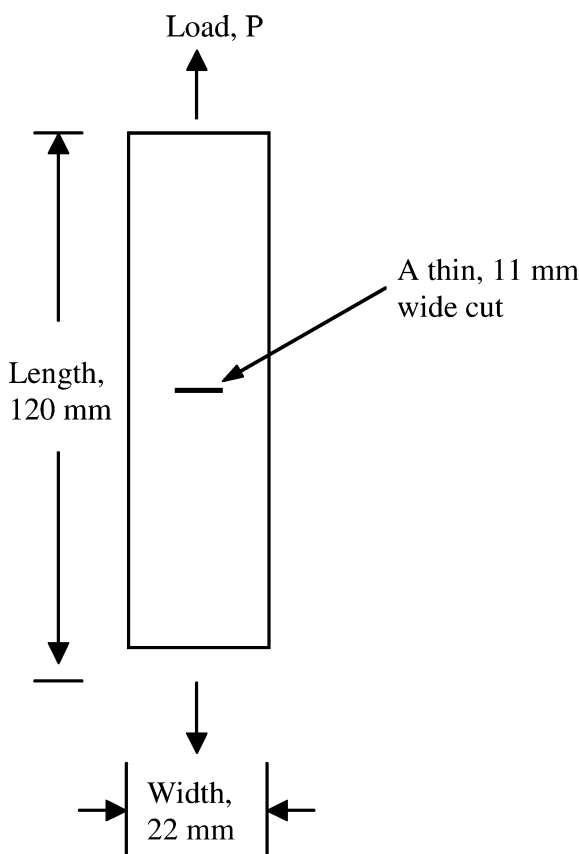
### Fracture surface energy measurement

A center-cracked tension (CCT) method was used to measure the fracture surface energy for linear elastic materials (Anderson 1995). The fracture surface energy is the energy required to fracture one unit area of material. The specimens were prepared by pouring the gelling solutions in between two smooth glass fiberboards uniformly spaced with 5 mm clearance. After at least 8 h, the PAAm gels were carefully removed from the assembly, and cut to the 5×22×120 mm shape for testing. Prior to analysis, the sample was cut with an 11 mm width notch in the middle of the specimen using a sharp blade (Fig. 1), and fixed with cyanoacrylate glue (Quik Tite, Loc Tite, Rocky Hill, CT) to the tension measurement fixture attached to an Instron 5540 universal testing machine (Instron Engineering Corp., Canton, MA) equipped with a 50 N load cell. The extension of all samples was at a constant speed of 2 mm/min, and all measurements were at room temperature. The calculation of fracture surface energy follows the equation (Zarzycki 1988),

$$2\Gamma = \frac{K_I^2(1 - \nu^2)}{E}, \quad (3)$$

where  $\Gamma$  is the fracture surface energy;  $K_I$  is the stress intensity factor;  $\nu$  the material Poisson's ratio, equaling to 0.5 for incompressible materials; and  $E$  the Young's modulus. The stress intensity factor  $K_I$  was calculated as (Anderson 1995),

$$K_I = \frac{P}{B\sqrt{W}} \sqrt{\frac{\pi a}{4W} \sec \frac{\pi a}{2W}} \left[ 1 - 0.025 \left( \frac{a}{W} \right)^2 + 0.06 \left( \frac{a}{W} \right)^4 \right], \quad (4)$$



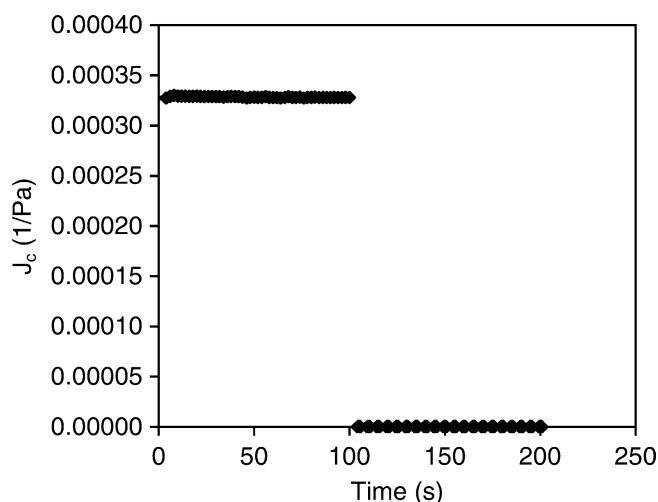
**Fig. 1** Center-cracked tension bar used for fracture surface energy determination

where  $P$  is the force of the specimen at failure;  $B$  is the specimen thickness;  $W$  is the specimen half width; and  $a$  is the half width of the notch.

## Results and discussion

### Elasticity of PAAm gels

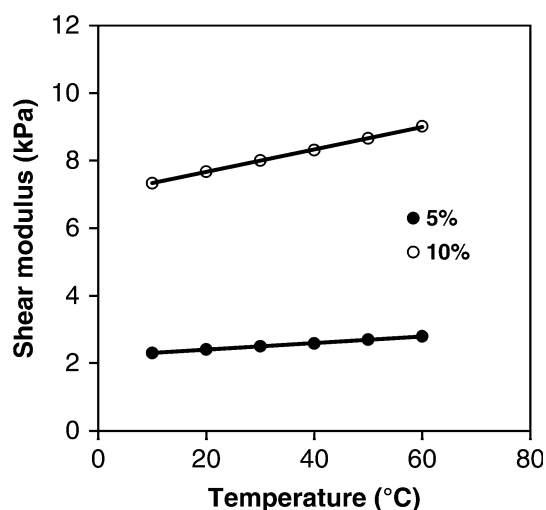
A creep recovery experiment was used to test the rheological properties of 5% PAAm gels with a monomer to crosslinker concentration ( $M/C$ ) ratio of 37.5. When a 200 Pa stress was applied for 100 s, a corresponding strain was produced simultaneously and remained constant (Fig. 2). Once the stress was removed, the strain immediately returned to zero, a typical creep recovery response for purely elastic solids. Another experiment to test the PAAm gel properties was the shear modulus dependence on temperature. The shear modulus changed proportionally with temperature at concentrations of 5% and 10% at a given  $M/C$  ratio of 37.5 (Fig. 3). The creep recovery test and the temperature dependence of shear modulus confirmed that polyacrylamide gels were elastic materials as stated by Foegeding et al.



**Fig. 2** Creep recovery of 5% PAAm gels ( $M/C$  37.5) at 25 °C

(1994), obeying the theory of rubber elasticity. Therefore, the network crosslink density (the number of crosslinks per unit volume) and the molecular weight between crosslinks could be calculated from shear modulus measurements according to Eqs. 1 and 2.

Most food gels are viscoelastic rather than elastic, and the theory of rubber elasticity cannot be strictly applied to these gels. However, many food gels do show substantial elasticity at certain concentrations and conditions, and Mitchell (1980) argued that if materials can be regarded as a cross-linked network, it may be possible to obtain the number of cross-links from the equilibrium shear modulus using the relationship purported by the theory of rubber elasticity. Segeren et al. (1974) found several features of the behavior of calcium alginate gels that were consistent with rubber elasticity theory. Also,

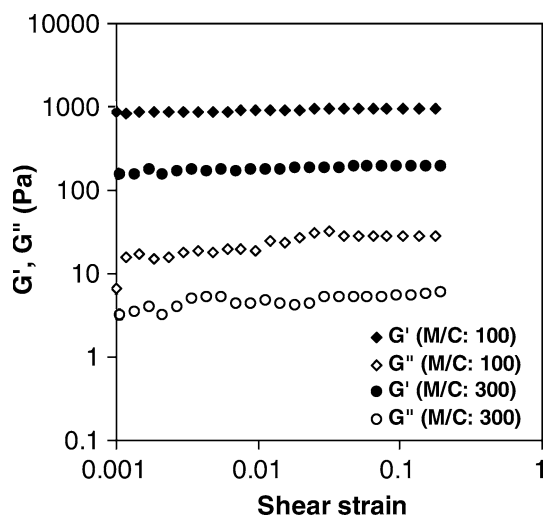


**Fig. 3** Shear modulus of PAAm gels ( $M/C$ : 37.5) versus temperature

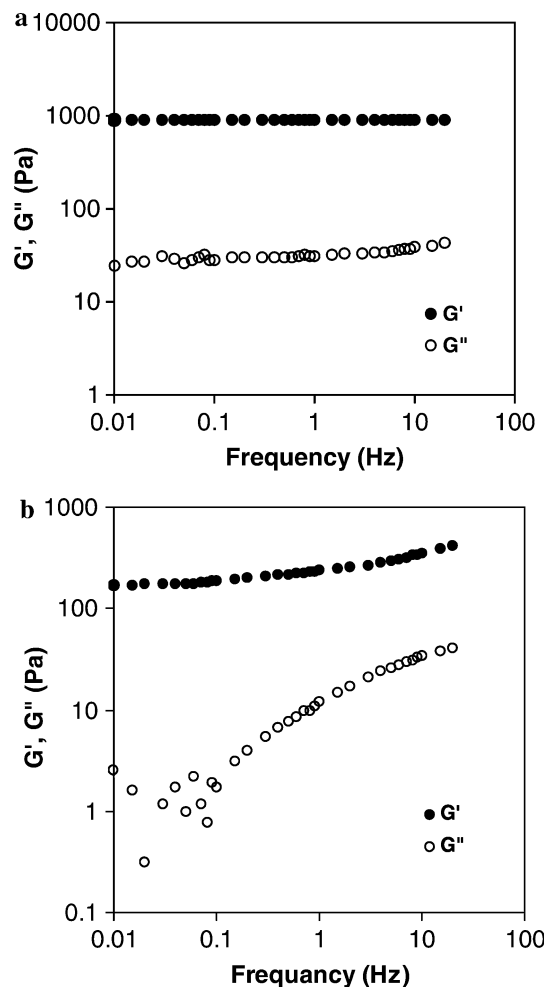
the properties of meat (Blanshard and Derbyshire 1975) and milk gels (Culioli and Sherman 1978) were discussed in terms of this theory. Furthermore, Van Kleef et al. (1978) first applied this theory to describe the nature and number of cross-links in heat-set protein gels, concluding that the theories of polymer physics could be used to relate the chemical structure of a protein gel to the physical properties. Oakenfull (1984) derived equations based on the theory of rubber elasticity to calculate, from measurements of shear modulus for non-covalent gels, the association constant for the formation of junction zones and the number of cross-linking loci.

#### Rheological properties of PAAm gels depending on gel composition

The mechanical spectra showed PAAm gels behaved viscoelastically when the monomer concentration was less than, or  $M/C$  ratio was more than some critical value. For instance, at a given  $M/C$  ratio of 37.5 and when the PAAm gel concentration was less than 2.4%, the gels displayed viscoelastic behavior. For 5% PAAm gels, when the  $M/C$  ratio was more than 200, the gels were viscoelastic. Figure 4 shows that for elastic and viscoelastic 5% PAAm gels,  $G'$  and  $G''$  remained constant within the range of strain accessible by the instrument, and the gel linear viscoelastic property was tested using torsional large deformation tests as shown and discussed later. A typical shear modulus-frequency relationship for elastic gels is shown in Fig. 5a, and a typical shear modulus-frequency relationship for viscoelastic gels is shown in Fig. 5b. Figure 5a shows the gel shear storage modulus  $G'$  was independent of frequency,



**Fig. 4** Representation of shear moduli associated with shear strain elastic ( $M/C$ : 100) and viscoelastic ( $M/C$ : 300) 5% PAAm gels at frequency of 0.1 Hz



**Fig. 5** a Representation of shear moduli associated with frequency for an elastic PAAm gel. The gel shown in this figure was 5% PAAm gel with  $M/C$  of 100 (25 °C). b Representation of shear moduli associated with frequency for a viscoelastic PAAm gel. The gel shown in this figure was 5% PAAm gel with  $M/C$  of 300 (25 °C)

indicating the gel was elastic. On the contrary, in Fig. 5b, the gel shear storage modulus  $G'$  changed with frequency, revealing that the gel tested was viscoelastic. The rheological response of PAAm gels with varying compositions are listed in Table 1, and these results indicate the rheological properties of PAAm gels could transit between elasticity and viscoelasticity depending on gel composition. Consequently, the  $M/C$  ratio should be in the range between 10 and 200 when investigating rheological properties of elastic PAAm gels.

The viscoelastic nature of PAAm gels can be explained considering structural characteristics. At a given  $M/C$  ratio, when the monomer concentration was low, the gel network was inhomogeneous (Naghsh et al. 1996). Accordingly, the gel permeability was high, resulting in viscous flow from solvent flowing within the matrix during deformation (van Vliet and Walstra



**Table 1** Rheological properties of PAAm gels

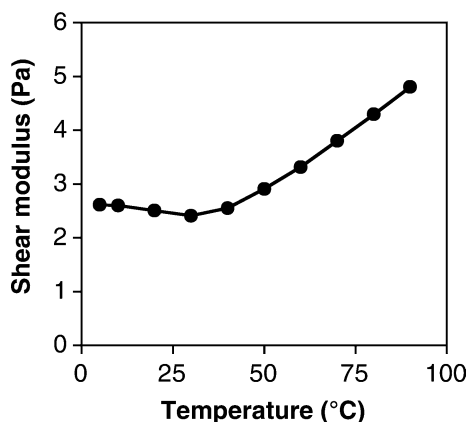
|                                 |     |     |     |     |     |      |      |      |      |      |      |
|---------------------------------|-----|-----|-----|-----|-----|------|------|------|------|------|------|
| $M/C$ (5% Monomer)              | 5   | 10  | 20  | 40  | 60  | 80   | 100  | 120  | 160  | 200  | 300  |
| Rheological property            | E   | E   | E   | E   | E   | E    | E    | E    | E    | VE   | VE   |
| Monomer conc. (%) ( $M/C$ 37.5) | 2.2 | 2.4 | 4.0 | 6.0 | 8.0 | 10.0 | 12.0 | 14.0 | 16.0 | 18.0 | 20.0 |
| Rheological property            | VE  | VE  | E   | E   | E   | E    | E    | E    | E    | E    | E    |

*E* Elastic; *VE* Viscoelastic.

1995). At a given monomer concentration, when the crosslinker concentration was low (high  $M/C$  ratio), the network chains were longer and more flexible to permit chain rearrangement with time during deformation, causing frequency-dependent behavior.

For elastic PAAm gels, the shear modulus is linearly proportional with temperature as demonstrated earlier, and the temperature dependence of stress for a constant strain has been established (James and Guth 1943). As expected, the shear modulus response of viscoelastic PAAm gels was different from elastic gels. Figure 6 shows the shear modulus change with temperature for 2.2% PAAm gels ( $M/C$ : 40). When the temperature was below 30 °C, the shear modulus decreased linearly with temperature, displaying an enthalpic property. However, when the temperature was above 30 °C, the shear modulus increased linearly with temperature, indicating an entropic property for the gels. As viscoelastic gels, the 2.2% PAAm gels were not fit to the theory of rubber elasticity.

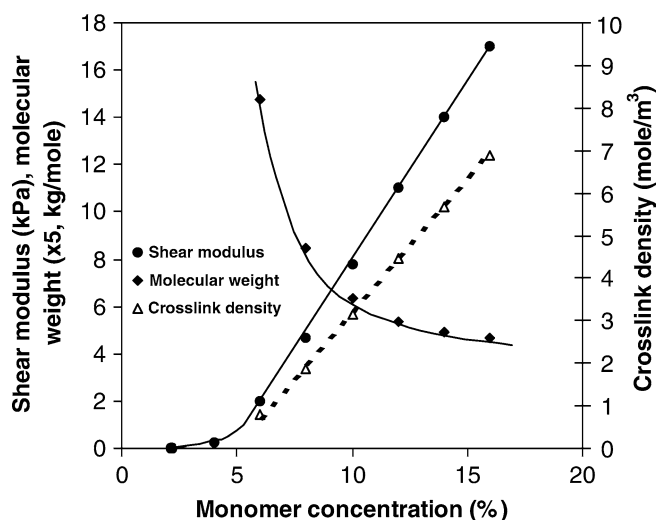
The 5% PAAm gels showed viscoelastic properties when the  $M/C$  ratio was more than 200 (Table 1). However, when the  $M/C$  ratio was less than 10, the gels were no longer transparent but opaque and brittle. The opacity in gels is known to be due to the spinodal decomposition of the polymer network, causing light scattering and transmission reduction for visible light (Durmaz and Okay 2001).



**Fig. 6** Temperature dependence of shear modulus for 2.2% PAAm gels ( $M/C$ : 37.5) at a 0.05 Hz frequency

Shear modulus associated with gel composition

Figure 7 shows the shear modulus associated with monomer concentration at a given  $M/C$  ratio of 40. A critical concentration of about 5% was observed to separate the curve into two regions. In the lower region, the shear modulus increased with monomer concentration slowly; whereas in the upper region, the shear modulus rapidly increased linearly with monomer concentration. An explanation for this phenomenon is that when the gel concentration was below 5%, the PAAm gels were inhomogeneous, with numerous clusters in which the crosslink density was higher than in other areas of the existing network (Naghsh et al. 1996). When the monomer concentration was above 5%, more uniform and compact gels formed. The crosslink density and the network chain molecular weight for gel concentration above 5% were calculated by taking  $g$  in Eq. (1) as 1.0 due to the incompressibility of PAAm gels. The network crosslink density was linearly proportional to the gel concentration, while the network chain molecular weight decreased nonlinearly with gel concentration. Consequently, if the gel fracture properties were studied in this concentration region, the relationship between network structure and the gel composition would be obtained, and this relationship would facilitate



**Fig. 7** Shear modulus associated with monomer concentration for PAAm gels ( $M/C$ : 40) at 25 °C

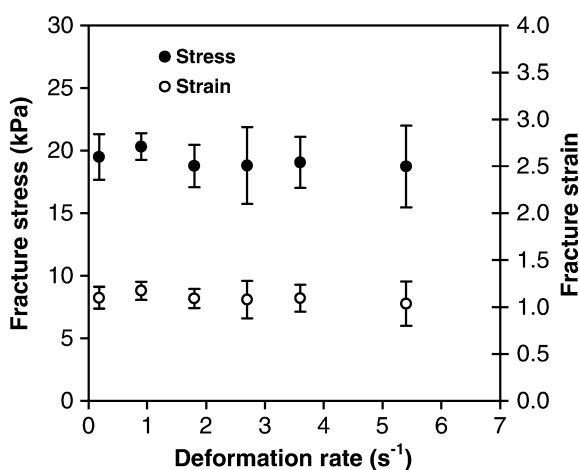
the analysis of the fracture properties and interpretation of fracture mechanisms.

#### The effect of strain rate on fracture properties

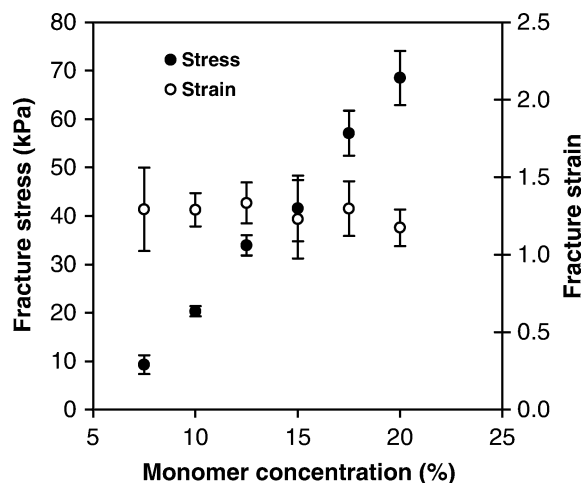
Figure 8 shows the fracture stress and strain of PAAm gels at different deformation rates in torsion, from 0.2 to 5.5 s<sup>-1</sup>. The fracture stress and strain were independent of deformation rate for PAAm gels, substantiating the elastic system claim. The fracture stress and strain values from large deformation tests reflect the energy and deformation required to cause fracture at a given deformation rate (Foegeding et al. 1994). For the elastic PAAm gels, all elastic energy released from the failed network chains was transferred to the neighboring chains as the energy for fracture propagation, resulting in deformation rate independence during fracture.

#### Fracture properties with respect to gel composition

Figure 9 shows the fracture stress and strain associated with monomer concentration at a given  $M/C$  ratio of 40. The monomer concentrations and the  $M/C$  ratio used here assured that all samples were elastic. The fracture stress increased linearly with gel monomer concentration, while the fracture strain remained constant. When gel concentration increased, the crosslink density increased as shown in Fig. 7, and accordingly, the gel strength increased, resulting in fracture stress increase. The fracture surface energy association with gel concentration is shown in Fig. 10. As mentioned earlier, the fracture surface energy is the energy to fracture one unit area of material; hence the fracture surface energy can be



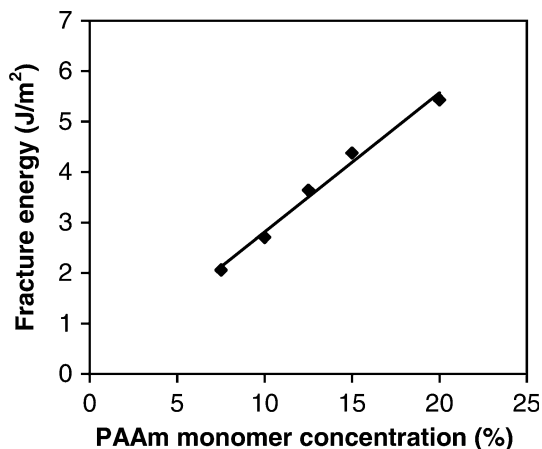
**Fig. 8** Fracture stress and strain for 10% PAAm gels ( $M/C$ : 40) associated with deformation rate in torsion (each point was the average of five samples, and the error bars show standard deviations)



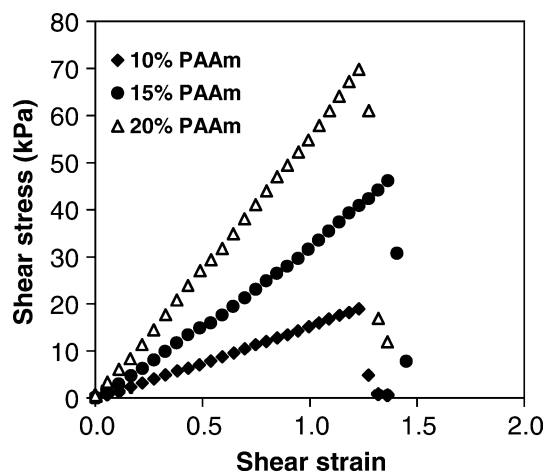
**Fig. 9** Fracture stress and strain for PAAm gels ( $M/C$ : 40) associated with gel concentration (each point was the average of five samples, and the error bars show standard deviation)

considered as a measurement for material cohesiveness. Figure 10 shows the gel fracture surface energy increased linearly with increasing gel concentration, indicating the cohesiveness correlated with the gel monomer concentration. Therefore, it can be purported that the fracture stress was the external force applied to overcome the gel cohesive force for PAAm gels.

Experimental observation showed the PAAm gels displayed a linear relationship between shear stress and shear strain when deformed to fracture for all elastic PAAm gels, and a representation for the stress-strain curves is shown in Fig. 11. The linear elastic behavior of the gels up to fracture indicated the linear stretching of the network chains, and the network chain length of the PAAm gels between crosslinks was such that the entire deformation process could be described by Gaussian statistics (as a random coil). Accordingly, the chains



**Fig. 10** Fracture surface energy of PAAm gels associated with gel concentration ( $M/C$ : 40) at room temperature



**Fig. 11** Shear stress versus shear strain of polyacrylamide gels in torsion ( $M/C$ : 40) at room temperature

between cross-links could be considered entropic springs. During deformation, the network chains became extended, and the force sustained by network chains was proportional to the extended length of the chains. When the force attained the largest force that the gel could sustain, the chain broke. Also the linear stress-strain relationship implied that the gel network chain did not experience finite chain extensibility; otherwise a strain-hardening behavior would result. Therefore the fracture mechanism of PAAm gels is different from those for engineering polymers, and cannot be explained by a nonlinear relationship of force and distance between polymer connections unlike gelatin (Groot et al. 1996).

The fracture shear strain of PAAm gels was approximately 1.3, which is considerably lower than that of rubber, but similar in magnitude to food gels. In comparison, the maximum extensibility for rubber corresponds with a shear strain of 10.0–20.0 (Treloar 1975). The small fracture strain and linear stress-strain relationship showed that the PAAm gels fractured in the linear region of the complete deformation curve. The fracture strain did not change with the network chain length, whose relation to monomer concentration was discussed during small strain tests (Fig. 7). This response also suggests that the fracture of PAAm gels was not a process of stretching network chains beyond their maximum extensibility, supporting a point that the fracture of PAAm gels occurred only when the stress magnitude sufficiently overcame the cohesive force binding the gel together.

The cause for a constant fracture strain cannot be established but could be related to the shortest network chain length. The fracture process was possibly a cascade course with accelerating speed (Bot et al. 1996), starting with a chain with the largest carrying stress. The chain with the highest carrying stress was normally the

shortest one in length. During deformation, the shortest chain broke first, and the energy released from the broken chain would transfer to the neighboring chains, accelerating the break of other chains. As a result, the fracture speed was so high that the gel fracture strain was mainly determined by the first broken chain. If the shortest chain length did not vary with concentration though an extreme wide variation in chain lengths for the whole network existed between the cross-links, then fracture strain would be constant.

The observed linear relationship of shear stress-strain up to fracture for PAAm gels at different monomer concentrations or  $M/C$  ratios indicates that the shear stress association with shear strain did not depend on the number of chains between crosslinks. Therefore, the results from PAAm gels in this study can be expanded to food gels, especially polysaccharide gels, which may have a varying number of statistical chain elements between cross-links.

The fracture stress increase with gel concentration is a general trend observed in many food gels. The constant strain at fracture, regardless of gel concentration, has also been observed for many food gels (Li et al. 1999; Mancini et al. 1999; Normand et al. 2000). The fracture stress of whey protein isolate gels increased with increasing protein concentration; whereas the fracture strain was insensitive to protein concentration when above 12% (Foegeding 1992). The increase of fracture stress with protein concentration promoted the increase of gel strength with concentration. Another example of the fracture property association with gel composition is egg white gels (Li et al. 1999). Fracture stress of egg white gels increased with protein concentration, while the fracture strains of the gels remained almost constant. The constant fracture strain as a function of concentration as observed for many food gels indicates that the fracture of these gels is not due to the length limitation of the network chains when they are stretched at deformation, but due to the incapability of the network to sustain large stresses, causing fracture prior to experiencing finite chain extensibility.

## Conclusions

The rheological properties of PAAm gels might be elastic or viscoelastic depending on gel network cross-link density. The fracture stress of PAAm gels correlated with monomer concentration, while the fracture strain remained constant regardless of the network chain length. The constant fracture strain coupled with the linear stress-strain behavior up to fracture indicated that the PAAm gel fracture did not result from the extensional limitation of the network chains, but rather resulted from the external force overcoming gel cohesiveness. Also, the linear stress-strain response up to



fracture for PAAm gels suggests, for gel systems, the simple stretching of network chains could not result in strain-hardening behavior. As a result, the theory of finite chain extensibility might not truly explain the strain-hardening behavior frequently observed for food polysaccharide gels.

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