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## Methods for evaluating the puncture and shear properties of pharmaceutical polymeric films

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### Summary

The tensile test is the most popular and widely used test for pharmaceutical films. From its data, the properties of Young's modulus, elongation to break, break strength and energy to break can be calculated. These properties though, do not reflect completely the behavior of a film when subjected to puncture and shear. The utility of puncture and shear data was evaluated by comparing it to data generated under tension for cast films of the following polymers: sodium carboxymethylcellulose, hydroxyethyl cellulose, hydroxypropyl methylcellulose and sodium alginate. Each test provided a different measure of elongation, strength and energy under the stress vs strain curve. For example, the puncture test had an enhanced ability to differentiate elongation. On the other hand, energy to shear was less than would be expected based on tensile and puncture data. Puncture and shear results provided an enhanced ability to interpret data and predict behavior of a polymer in its finished application.

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### Introduction

Traditionally, stress-strain testing in the tensile mode has been the most popular and widely used mechanical test for pharmaceutical films. The tensile test is practical, and analysis of its data is relatively straightforward. For example, it has been used to study the effects of humidity (Aulton, 1982), plasticizer (Aulton et al., 1981), solid filler (Aulton et al., 1984; Okhamafe and York, 1985), and method of manufacturer (Vemba et al., 1980) on the tensile properties of polymeric film coat-

ings for tablets. The tensile test not only gives an indication of elasticity and strength but also of toughness. However, polymers are viscoelastic and their mechanical behavior is dependent upon many factors. Consequently, the tensile test is often only a rough guide to the behavior of a polymer in its finished application. In particular, this can be illustrated by finished applications where film-coated particles are subjected to puncture and shear as in the processes of compression and chewing.

Because puncture and shear properties are not directly obtained from tensile parameters such as Young's modulus and ultimate tensile strength, two new devices are presented in this report that examine directly the processes of puncture and

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shear on pharmaceutical films. Both devices are designed to use the drive and force detecting mechanisms of a stress-strain tester. The first device measures resistance to deformation with a puncturing probe. Stress is measured as a function of probe displacement. The second device measures resistance to shear by a punch-type shearing tool. Shear strength is determined by measuring stress as a function of punch displacement. We evaluated the utility of the two new devices by comparing data generated under puncture and shear with data generated under tension on a series of cast polymeric films with significantly different degrees of elasticity, strength and toughness. The polymers examined were sodium carboxymethylcellulose, hydroxyethyl cellulose, hydroxypropyl methylcellulose and sodium alginate. It was found that normal intuitive interpretations of tensile data are inadequate for predicting puncture and shear properties.

## Materials and Methods

### Materials

Sodium carboxymethylcellulose (Grade 7H4F) and hydroxyethyl cellulose (Natrosol 250L) were obtained from Hercules, Inc., Wilmington, DE. Hydroxypropyl methylcellulose (Methocel E4M) was obtained from Dow Chemical, Inc., Midland, MI, and sodium alginate (Kelgin XL) was obtained from Kelco, Inc., Chicago, IL.

### Film preparation

Aqueous solutions of each polymer were carefully poured to prevent bubble formation, onto clean  $20 \times 20$  cm glass plates. Barriers were fitted on the edges of the plates to prevent overflow. The amount of solution poured was such that two grams of polymer would remain on the plate after the water evaporated. Dried films were removed from the glass plates. Films for tensile tests were punch-cut (Punch-Press, Model 8463, MS Instrument Co., Stony Creek, NY) into  $15.24 \times 2.54$  cm strips. Films for puncture and shear tests,  $7.5 \times 7.5$  cm squares, were cut with scissors. All cut films were conditioned at  $22^\circ\text{C}$  and 50% relative humidity for a minimum of 96 h prior to testing.

The thickness of each cut film was measured with an electronic digital readout gauge (Starret Electronic Height Gauge, L.S. Starret Co., Athol, MA).

### Apparatus

The stress-strain tester used for all studies was an Instron Model 1000, fitted with a 5 or 50 kg load detecting transducer (Instron Corp., Canton, MA). Load vs displacement data were acquired and transformed to stress vs strain data using modified proprietary software (IMAP 2.3, Systems Integrated Technology, Inc., Stoughton, MA) and a personal computer with 640K RAM (IBM PC/XT). The puncture and shear test devices were custom-designed, and are schematically illustrated in Figs. 1–3. Each device was made of hardened steel.

### Test procedures

All tests were conducted at  $22^\circ\text{C}$ . For tensile tests, film specimens were clamped with 2.54 cm pneumatic grips. The rate of strain was 10 mm/min. Load vs displacement data were recorded until the specimen broke. This data was then converted to stress vs strain. Young's modulus, elongation to break, break strength, and energy to break were calculated for each specimen.

For puncture tests, the puncturing probe was attached to the drive mechanism of the stress-strain tester. The film holder (Fig. 1) was mounted such that the center of the cylindrical hole was directly below the drive path of the puncturing probe (Fig. 2). Cut films were positioned between the two mounting plates and holding screws were tightened sufficiently to prevent slippage of the specimen. At a velocity of 10 mm/min, the hemispherical end ( $r = 0.475$  cm) of the puncturing probe was driven down through the mounted film. Load vs displacement data were recorded from the point of contact of the probe with the film until the film was ruptured. These data were then converted to stress vs strain. Elongation to puncture, puncture strength and energy to puncture were calculated for each film.

For shear tests, the shearing tool was also attached to the drive mechanism of the stress-strain

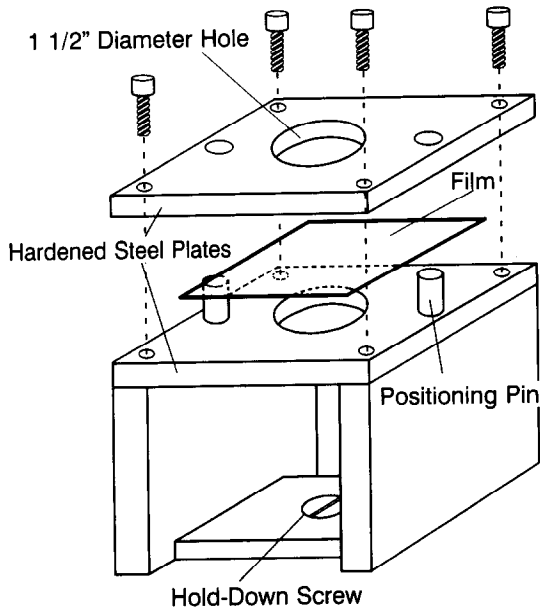


Fig. 1. Exploded 3-dimensional diagram of the holder used to mount films for puncture and shear testing.

tester. The film holder (Fig. 1) was mounted such that the center of the cylindrical hole was directly below the drive path of the shearing tool (Fig. 3). Prior to mounting the film specimen, a 0.125 cm

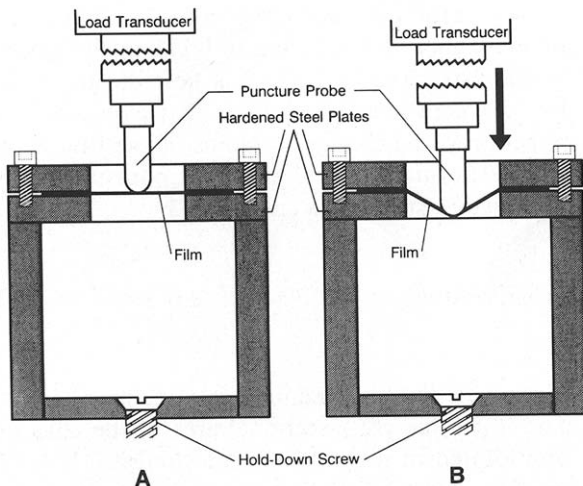


Fig. 2. Cross-sectional diagram of a mounted film for puncture testing with a hemispherical probe. A: the position of the puncturing probe just prior to deformation of the film. B: the position of the puncturing probe after the film has elongated, but prior to puncture.

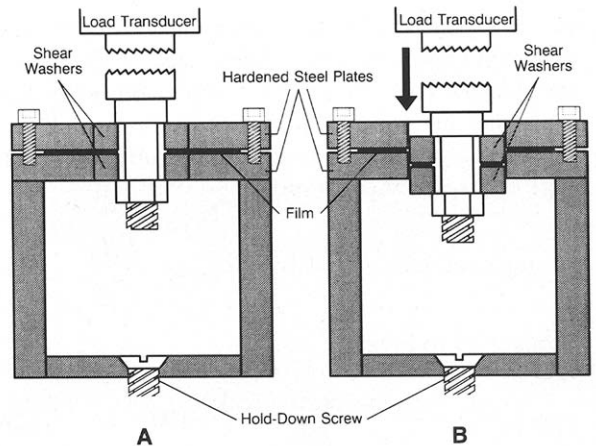


Fig. 3. Cross-sectional diagram of a mounted film for shear testing. A: the position of the shearing washers prior to initiation of shear. B: the position of the shear washers after completion of shear. Clearance between the shear washers and film holder is 0.038–0.064 mm.

diameter hole was punched in the center of each specimen. The specimen was then mounted with its punched hole in the center of the cylindrical hole of the film holder. With one of the washers positioned on top of the film, the threaded shaft of the shearing tool was lowered through the hole in the washer and film. The lower washer was then positioned in contact with the lower surface of the film and a hex nut was tightened by hand to hold the two washers in position. At a velocity of 1.0 mm/min, the shearing tool was driven down through the mounted film. Load versus displacement data were recorded from the time of initial movement of the shearing tool until the film was sheared around the complete circumference of the cylindrical hole. The data were then converted to stress vs strain. Shear strength and energy to shear were calculated for each film.

## Theory

### Tensile tests

Typical stress-strain curves and the theory behind the calculation of Young's modulus, elongation to break, break strength and energy to break for stress-strain tests in the tensile mode are well documented (Aulton, 1982; Shah, 1984) and will

not be repeated here. It is necessary to present the resultant expressions that define each of the above parameters for convenient comparison with expressions derived for puncture and shear tests. The expressions assume data is acquired as load (kg) vs displacement (mm).

$$\text{Young's modulus} = E' \text{ (dyne/cm}^2\text{)} = \frac{S_0 L_0}{A_R} \quad (1)$$

Elongation to break

$$= \epsilon_B (\%) = \frac{\text{extension to break}}{L_0} \cdot 100 \quad (2)$$

$$\text{Break strength} = B.S. \text{ (dyne/cm}^2\text{)} = \frac{F}{A_R} \quad (3)$$

Energy to break per unit volume

$$= \Delta E_B \text{ (erg/cm}^3\text{)} = \frac{AUC}{V_0} \quad (4)$$

where  $S_0$  is the initial slope of the linear portion of the stress-strain curve;  $L_0$  is the original length of the sample between the clamps;  $A_R$  is the rectangular cross-sectional area of the specimen;  $F$  is the load required to break the specimen;  $AUC$  is the area under the load vs displacement curve;  $V_0$  is the volume of the specimen between the clamps and is equal to  $A_R \times L_0$ .

#### Puncture tests

A typical plot of acquired load vs displacement data for a film subjected to puncture is shown in Fig. 4. The parameters of most significance are the displacement of the probe from initial contact to puncture of the film, area under the curve and peak load. From this data, elongation to puncture, puncture strength and energy to puncture were calculated. The nature of the test did not allow calculation of Young's modulus. Elongation to puncture is calculated by:

Elongation to puncture

$$= \epsilon_p (\%) = \frac{([R]^2 + [D]^2)^{1/2} - R}{R} \cdot 100 \quad (5)$$

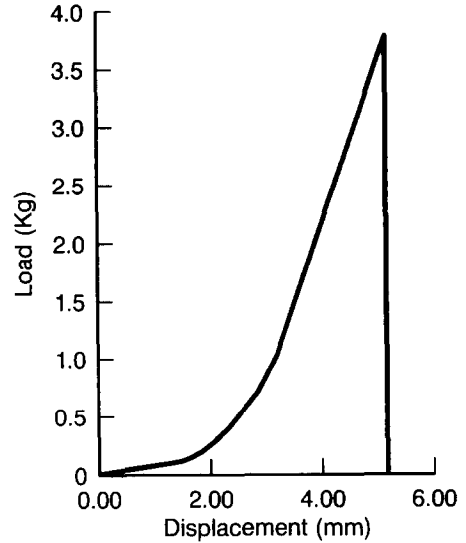


Fig. 4. Typical plot of acquired load vs displacement data for films subjected to puncture. The height of the peak is the load required to puncture the film.

where  $R$  is the radius of the film exposed in the cylindrical hole of the film holder; and  $D$  is displacement of the probe from point of contact to point of film puncture. It is important to note that elongation to puncture is not calculated in the same manner as elongation to break in the tensile mode. The basis of Eqn. 5 is best illustrated in Fig. 2 where it becomes clear that elongation to puncture is the change in radius of the film from its predeformation state to that at puncture.

Puncture strength is calculated by:

$$\text{puncture strength} = P.S. \text{ (dyne/cm}^2\text{)} = \frac{F}{A_{cs}} \quad (6)$$

where  $F$  is the load required to puncture the film and  $A_{cs}$  is the cross-sectional area of the edge of film located in the path of the cylindrical hole of the film holder. Division by  $A_{cs}$  normalizes the data for differences in thickness from film to film.

The equation for energy to puncture is similar to that for the tensile test, except for the calculation of the volume term. Hence,

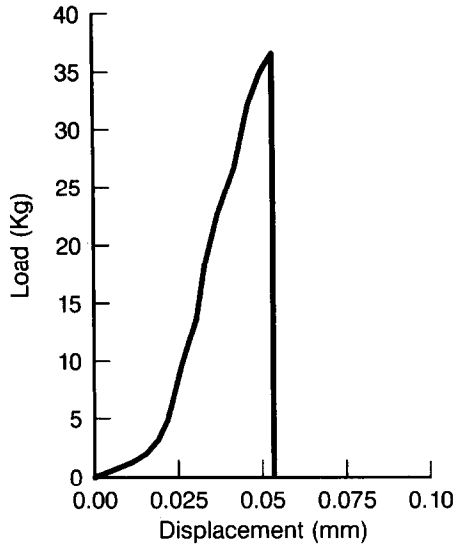


Fig. 5. Typical plot of acquired load vs displacement data for films subjected to shear. The height of the peak is the load required to shear the film.

Energy to puncture per unit volume

$$= \Delta E_p (\text{erg/cm}^3) = \frac{AUC}{V_c} \quad (7)$$

where  $V_c$  is the volume of the film located in the die cavity of the film holder.

#### Shear tests

A typical plot of acquired load vs displacement data for a film subjected to shear is shown in Fig. 5. Significant features of the plot are area under the curve and peak load. From this data, shear strength and energy to shear were calculated. The nature of the test did not permit calculation of Young's modulus or elongation to shear. Shear strength is calculated by:

$$\text{shear strength} = S.S. (\text{dyne/cm}^2) = \frac{F}{A_{cs}} \quad (8)$$

where  $F$  is the load required to shear the specimen and  $A_{cs}$  is as defined previously. The equation for energy to shear is as follows:

energy to shear per unit volume

$$= \Delta E_s (\text{erg/cm}^3) = \frac{AUC}{V_c} \quad (9)$$

where  $AUC$  and  $V_c$  are as previously defined.

## Results and Discussion

### Interpretation of tensile data

Characteristic features of stress-strain curves of tensile tests are routinely used to characterize polymer properties (Billmeyer, 1984). For example: a soft and weak polymer is characterized by low modulus, low ultimate tensile strength and low elongation to break; a hard and brittle polymer is characterized by high modulus, moderate ultimate tensile strength and low elongation to break; a soft and tough polymer is characterized by low modulus, moderate ultimate tensile strength and high elongation to break; and a hard and tough polymer is characterized by high modulus, high ultimate tensile strength and high elongation to break. Toughness is directly proportional to area under the stress-strain curve which is quantitated as energy. The greater the amount of energy a polymer can absorb prior to break, the greater its toughness.

In the case where polymer coatings are applied to particles that will be compressed into tablets, it is desirable to have coatings that are resistant to damage caused by compression. In addition, if the compressed tablet is chewable, it is desirable to have coatings that are resistant to damage caused by chewing. Therefore the coatings need to withstand common forms of deformation such as puncture and shear. Hence, if tensile data alone were used to predict puncture and shear properties, then one might expect hard and tough polymers to best withstand the rigors of compression and chewing. From the data in Table 1, where sodium alginate and sodium carboxymethylcellulose films are compared to hydroxyethyl cellulose and hydroxypropyl methylcellulose films, sodium alginate films and sodium carboxymethyl cellulose films are hard and tough, while hydroxyethyl cellulose films followed by hydroxypropyl methylcel-

TABLE 1

Results of tensile tests at a rate of strain of 10 mm/min, at 22°C, where *n* is the number of film specimens tested

Polymer	Film thickness (mm)	Young's modulus $\times 10^{10}$ (dyne/cm <sup>2</sup> )	Elongation to break (%)	Break strength $\times 10^8$ (dyne/cm <sup>2</sup> )	Energy to break $\times 10^7$ (erg/cm <sup>3</sup> )
NaCMC <sup>a</sup>	0.038	5.03	4.11	11.14	3.09
( <i>n</i> = 8)	0.004	0.38	0.99	1.46	1.12
HEC <sup>b</sup>	0.041	0.72	17.06	3.51	4.15
( <i>n</i> = 8)	0.004	0.06	4.11	0.30	1.24
HPMC <sup>c</sup>	0.042	2.18	10.35	8.19	6.10
( <i>n</i> = 8)	0.003	0.23	1.06	0.19	0.69
NaA <sup>d</sup>	0.035	5.54	4.96	13.04	4.47
( <i>n</i> = 8)	0.001	0.23	0.53	0.57	0.68

Values are means and S.D.

<sup>a</sup> Sodium carboxymethylcellulose; batch B-174.

<sup>b</sup> Hydroxyethyl cellulose; Batch B-176.

<sup>c</sup> Hydroxypropyl methylcellulose; batch B-178.

<sup>d</sup> Sodium alginate; batch B-175.

lulose films are soft and tough. Consequently, sodium carboxymethylcellulose films and sodium alginate films would be expected to withstand puncture and shear better than hydroxypropyl methylcellulose films or hydroxyethyl cellulose films.

#### Interpretation of puncture data

In general, the puncture properties of polymeric films are related to toughness. As with the case of tensile data, toughness is directly proportional to the area under the stress versus strain curve and is quantitated as energy. Puncture strength is a measure of toughness and is directly proportional to resistance to break or fracture. Based on the data in Table 2, the hydroxypropyl methylcellulose films required the greatest amount of energy to puncture, while sodium alginate films required the lowest. Similarly, hydroxypropyl methylcellulose films had the highest puncture strength, while sodium alginate films had the lowest. In addition, sodium alginate films have the lowest elongation to break. Therefore, sodium alginate films are least resistant to puncture.

The polymer films that are most resistant to puncture are hydroxypropyl methylcellulose films. This results from their high energy to puncture, high puncture strength and relatively high elonga-

tion to puncture. The films with the next highest resistance to puncture are those made of sodium carboxymethylcellulose. These films have the next highest combination of puncture strength and energy to puncture. It should be noted though, that the elongation to puncture of sodium carboxymethyl cellulose films is relatively low. On the other hand, hydroxyethyl cellulose films show the highest elongation to puncture and the lowest puncture strength.

TABLE 2

Results of puncture tests at a rate of puncture of 10 mm/min, at 22°C where *n* is the number of film specimens tested

Polymer	Film thickness (mm)	Elongation to puncture (%)	Puncture strength $\times 10^8$ (dyne/cm <sup>2</sup> )	Energy to puncture $\times 10^7$ (erg/cm <sup>3</sup> )
NaCMC	0.037	0.55	1.10	1.65
( <i>n</i> = 7)	0.003	0.21	0.15	0.39
HEC	0.039	2.52	0.64	1.70
( <i>n</i> = 9)	0.004	1.13	0.15	0.64
HPMC	0.039	1.80	1.16	2.63
( <i>n</i> = 8)	0.003	0.64	0.20	0.77
NaA	0.034	0.24	0.80	0.97
( <i>n</i> = 6)	0.002	0.15	0.17	0.39

Values are means and S.D.

TABLE 3

Results of shear tests at a rate of shear of 1.0 mm/min, at 22°C where  $n$  is the number of film specimens tested

Polymer	Film thickness (mm)	Shear strength $\times 10^8$ (dyne/cm <sup>2</sup> )	Energy to shear $\times 10^7$ (erg/cm <sup>3</sup> )
NaCMC	0.037	5.31	0.23
( $n = 7$ )	0.002	1.49	0.25
HEC	0.040	2.63	3.72
( $n = 8$ )	0.003	0.66	1.24
HPMC	0.039	6.05	3.47
( $n = 8$ )	0.003	1.22	2.42
NaA	0.034	7.36	7.86
( $n = 7$ )	0.002	1.22	2.55

Values are means and S.D.

#### Interpretation of shear data

Shear strength is defined as the ability to withstand the maximum load required to shear the specimen so that the moving portion completely clears the stationary portion. Coated particles undergoing compression or chewing are likely to experience shear in addition to puncture deformation. The shear test is useful for very thin specimens that may stretch excessively and give misleading results when tested in the tensile mode. Generally, shear strength is on the order of one-half the break strength.

Examination of shear data in Table 3 shows sodium alginate films to have the highest shear strength and energy to shear. Consequently, sodium alginate films are the most superior of the films examined in this study for resisting shear. It should also be noted that they are the most inferior films for resisting puncture. Hence, it is important to realize that a given test can emphasize one material property over another. For example, the shear test emphasizes strength, while the puncture test emphasizes elongation. Even so, note that elongation does occur during the shear test prior to shear, and complete shear of the film is not instantaneous. If complete shear were instantaneous, there would be no area under the stress vs strain curve, and energy to shear could not be calculated by Eqn. 9.

#### Comparison of tensile, puncture and shear test methods

Examination of the data in Tables 1–3 shows that puncture and shear testing augments tensile testing. As can be expected, the absolute values of energy, strength and elongation for each test and each polymer are of different magnitude. One approach to quick comparative analysis is the use of ratios determined by using the values obtained for one polymer, such as sodium alginate, as the common denominator. For example, when elongation results for the tensile and puncture tests are compared (Table 4), the tensile test shows that hydroxyethyl cellulose films stretch about 3.5 times more than sodium alginate films, but the puncture test shows a 10.5 fold difference. Hence, the puncture test provides greater differentiation of the elongation property and could be useful to differentiate films the tensile test may show comparable. This does not suggest that the puncture test should replace the tensile test, but the tests should complement each other.

Likewise, measurements via one test method should not be interpreted in isolation. For example, the magnitude of energy to break, shear or puncture is determined by the contributions of strength and elongation. When tested in tensile mode, hydroxyethyl cellulose films and sodium alginate films have comparable energies to break (Table 5), but the break strength of sodium alginate is about 4 times that of hydroxyethyl cellulose (Table 6), and as previously stated the elongation of hydroxyethyl is about 3.5 times that of sodium alginate. Hence, the energy to break sodium alginate films comes primarily from break strength,

TABLE 4

Ratio obtained by dividing the experimentally obtained value of elongation to break and elongation to puncture for each polymer film by the corresponding experimentally obtained value for sodium alginate

Polymer	Tensile test	Puncture test
NaCMC	0.83	2.29
HEC	3.44	10.5
HPMC	2.09	7.5
NaA	1.0	1.0

TABLE 5

Ratio obtained by dividing the experimentally obtained value of energy to break, energy to puncture and energy to shear for each polymer film by the corresponding experimentally obtained value for sodium alginate

Polymer	Tensile test	Puncture test	Shear test
NaCMC	0.69	1.70	0.03
HEC	0.93	1.75	0.47
HPMC	1.36	2.71	0.44
NaA	1.0	1.0	1.0

while the energy to break hydroxyethyl cellulose films comes primarily from elongation. In the puncture mode, the energy to puncture hydroxyethyl cellulose films is about 1.75 times that of sodium alginate films (Table 5), but the puncture strength is about 0.8 (Table 6) and elongation to puncture is about 10.5 times greater (Table 4). Hence, the puncture mode of testing highlighted the contribution of elongation, to energy of the film to fail.

When one test is used alone, erroneous extrapolation to behavior in its finished application is more likely. For example, if only tensile data were available for interpretation, one might intuitively expect a polymer film with high elongation to break to best withstand the rigors of chewing and compression when it is coated onto particles. But in chewing and compression, films coated on particles are subjected to puncture and shear. Although films of sodium carboxymethylcellulose,

TABLE 6

Ratio obtained by dividing the experimentally obtained value of break strength, puncture strength and shear strength for each polymer by the corresponding experimentally obtained value for sodium alginate

Polymer	Tensile test	Puncture test	Shear test
NaCMC	0.85	1.38	0.72
HEC	0.27	0.80	0.36
HPMC	0.63	1.45	0.82
NaA	1.0	1.0	1.0

hydroxyethyl cellulose and hydroxypropyl methylcellulose have elongations to puncture greater than sodium alginate (Table 4), each has a lower shear strength (Table 6) and lower energy to shear. Therefore, shear data when used in conjunction with tensile and puncture data enhance one's ability to interpret data and predict the behavior of a polymer in its finished application.

Puncture and shear testing should be used in addition to tensile testing in evaluating polymer films. Each test provides a different measure of elongation, ultimate strength and energy. These tests are relatively easy to perform and can be adapted to accommodate conventional stress-strain testing equipment.

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