# The Effects of Inclusions and Conditions of Storage on the Mechanical Properties of Maize Starch and Methylcellulose Films

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Abstract—Films of methylcellulose and gelatinized maize starch were tested in tension, with and without 0.01-10% of additive in the film. The effects of these inclusions on the physical properties of the films have been found to be statistically significant in almost every case. The films, with and without inclusions, have also been conditioned at four different relative humidities and the behaviour of these films has also been considered. In general films were weakened by the presence of additives particularly at higher concentrations. The properties of methylcellulose films were changed most at specific concentrations of the inclusions, this effect was not found with maize starch films. Plasticization of maize starch films was not achieved.

In many cases the manufacture of tablets involves the process of wet massing and screening and the inclusion of a filmforming binder. The binder confers strength to the resulting granules and often improves the quality of the final tablets. The mechanical properties of films formed using tablet binders have been studied using pure binders (Healey et al 1974; Reading & Spring 1984), and with inclusions (Reading & Spring 1985). Polymeric films suitable for film coating have also been studied with plasticizers (Aulton 1982; Rowe 1982). During the wet massing and screening operation some of the components of the tablet formulation will dissolve in the binder fluid and then be deposited in the film of binder when the granules are dried. Wells et al (1982) have reported an improvement in the mechanical properties of tablets due to the inclusion of plasticizers in tablets. The effects on the mechanical properties of secondary materials dissolved in the binder film may well be significant for the tableting process and so some potential plasticizers and the tablet diluent, lactose, have been studied for their effects on binder films.

A plasticizer has been defined by Bernardo & Burrell (1972), as a low molecular weight compound of low volatility which when added to another material changes the physical and chemical properties in such a manner that the finished product is in a more useful form. A plasticizer therefore, serves to alter such physical properties as flexibility, hardness, tensile strength, and elasticity. The way this function is performed depends on the unique property of the plasticizer to form a combination/interaction with the polymer which normally occurs at the molecular level with some overlap at the macromolecular level. Polymer-plasticizer systems differ widely and compatibility is specific, that is to say, that if a plasticizer is compatible with one type of polymer it does not necessarily ensure compatibility with another polymer. Generally, compatibility is thought of as an indication of solubility in which the plasticizer behaves as a solvent for the polymer. Compatibility is defined as the amount of the plasticizer that can be added before phase separation occurs.

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Antiplasticizers are materials which exert an opposite effect to plasticizers on the polymer and this is usually indicated by an increase in tensile strength and Young's modulus, and a decrease in percentage elongation at fracture.

The presence of solid inclusions tends to have the same effect as antiplasticizers. Solid inclusions decrease the ultimate tensile strength and percentage elongation at fracture but increase the modulus if elasticity and with some polymers, e.g., hydroxypropyl methylcellulose, actually render them harder and more brittle.

## **Materials and Methods**

Films were prepared using methylcellulose (Methocel A15, Colorcon Ltd., Orpington, UK) and maize starch (BDH Chemicals Ltd, Poole, UK). The inclusions were lactose, polyethylene glycol 400 & 600 and propylene glycol (all from BDH Chemicals Ltd).

## Preparation of films

Methylcellulose. Films were prepared by casting a 7.5% w/w aqueous solution of binder onto clean glass plates using a chromotography spreader. Films were dried at  $45 \pm 2^{\circ}$ C in an oven for  $1-1\frac{1}{2}$  h to a moisture level that permitted coherent films to be cut and removed. The dried film was cut into strips  $5 \times 0.5$  cm along the plane of spreading.

Maize starch. A 7.5% w/w paste of maize starch gelatinized and spread at 90°C was used to prepare films. The glass plates used were pretreated with dichlorodimethyl silane, allowed to air dry, rinsed thoroughly with distilled water and dried at 60°C. The spread films were dried and cut as for methylcellulose.

The films thicknesses ranged from  $40-55 \ \mu$ m, films with variations in thickness greater than  $5 \ \mu$ m along the length or any visible defects were rejected. The strips were then conditioned for 7 days in glass desiccators containing saturated salt solutions to give the required humidity (Winston & Bates 1960; Merck Index 1968), the desiccators

were stored in a constant temperature cabinet at 25°C. This length of time has been found adequate for equilibration of films (Reading 1984). The films were tested as described by Reading & Spring (1984). Ten strips were tested for each set of parameters studied.

## **Results and Discussion**

## Inclusions in methylcellulose films

The effects of inclusions on the mechanical properties of methylcellulose films in tension at different relative humidities are given in Tables 1–4. The values in the Tables are the mean of ten replicate measurements, these means had coefficients of variation of 9–16%. Despite this variability, analyses of variance, Table 5, show that in almost every case the inclusion, the relative humidity and their mutual interaction have a significant effect on the mechanical properties of the films.

Effects due to lactose (Table 1). The analyses of variance indicate that lactose concentration had a significant effect on all the measured properties. It can be seen from Table 1 that for films stored at 12, 44 and 65% r.h. the values for all the properties peak at 0.5% lactose, except for proportional limit (the load at which the linear part of the load-extension curve ends, expressed as a percentage of the load at failure) where the highest values are at 5% inclusion. At 81% r.h. the differences between the values for 0.2, 0.5 and 1% inclusion were much less marked, but were still higher than the values at lower and higher concentrations. It is clear also for Young's modulus, elastic resilience, and proportional limit, that the relative humidity had a much greater effect than the additive on film properties. The values obtained for these properties were significantly higher for films conditioned at 44% r.h. The presence of 0.5% lactose in the film had a strengthening effect but at higher concentrations of lactose the films were weakened. Visual inspection showed that lactose crystallized from the film at the higher concentrations especially at 81% r.h.

Effects due to PEG 400 (Table 2). PEG 400 concentration had a significant effect on all the properties measured. Table 2 shows that those values tend to a minimum at 0.2% PEG 400, except for the proportional limit. Above 1%, additive values for tensile strength, Young's modulus, and elastic resilience, fell, whereas those for elongation at fracture rose. As with lactose, there is a concentration at which film properties are altered to a greater extent than at higher and lower concentrations and PEG 400 serves to weaken the film and plasticize it at concentrations above 1%.

Effects due to PEG 600 (Table 3). The effects due to PEG 600 were essentially the same as those for PEG 400 except for a greater tendency for the values for Young's modulus to peak at 0.5% additive in the film. The results obtained did not show significant differences due to relative humidity for toughness and elongation at fracture.

The two polyethylene glycols tested showed classic plasticization above 1% in the film. Below 1%, the minima at 0.2%for many of the films tested is less easy to explain. The films were clearly weakened and also showed a reduction in elongation at fracture; the latter would be expected with antiplasticization, which has been reported for low plasticizer concentrations (Jackson & Caldwell 1965), but antiplasticization should also give increased tensile strength and Young's modulus. That effect was not present.

Table 1. Mechanical properties of methylcellulose films. Values represent the mean of 10 replicate measurements. Inclusion: lactose

		Percentage inclusion							
	r.h.	0	0.01	0.1	0.2	0.5	1	5	10
Ultimate tensile	12%	53.6	<b>4</b> 8·7	44·8	45.6	59·4	47.4	45.9	44.(
strength $Nm^{-2} \times 10^{6}$	44%	55.5	47.5	<b>48</b> ∙0	46.2	54.8	50.6	47·8	44.0
	65%	55.5	53.1	<b>48</b> ∙4	<b>49</b> ·5	56.6	46.4	<b>46</b> ∙0	47.7
	81%	<b>41</b> ·4	50.9	48·9	54·9	54.5	<b>4</b> 4·5	41·I	37.4
Toughness $Jm^{-3} \times 10^5$	12%	100	90.8	86.9	83·0	114.8	93·2	80-9	77.7
	44%	92.9	73.6	85.6	64·2	111.2	76·2	64·7	60.8
	65%	105.7	104·0	84.6	82·1	103.7	83·2	62.1	75.4
	81%	88·3	<b>98</b> ∙7	<b>96</b> ∙2	113.6	111.7	75·2	90.6	63·2
Young's modulus	12%	124.6	112.9	100-9	115.6	121.4	111.8	114.6	103-0
$Nm^{-2} \times 10^{7}$	44%	143·0	129.6	123.1	140.8	143.7	130·2	130.0	121.8
	65%	111-5	103-1	101·4	98·4	108-3	93.6	96·7	88.7
	81%	<b>89</b> ∙6	<b>9</b> 7·7	92·5	102.5	104·0	91·1	95·2	70· <del>(</del>
Elastic resilience	12%	3.4	2.9	2.4	1.8	3.6	2.1	2.8	2.8
$Jm^{-3} \times 10^{5}$	44%	4.6	3.7	3.8	3.4	4∙6	<b>4</b> ∙0	4·2	3.8
	65%	3.4	2.7	2.3	2.4	3.2	2.4	2.7	3.(
	81%	1.8	1.9	2.0	1.8	1.9	1.9	1.8	1.8
Elongation at	12%	46.5	<b>47</b> ∙0	46.9	<b>46</b> ∙8	<b>48</b> ·4	45-4	39.0	31.7
fracture %	44%	<b>46</b> ∙0	36.9	43·8	32.5	50·9	36.7	31-8	29.(
	65%	46.7	<b>50</b> ∙0	42·0	<b>40</b> ·7	<b>4</b> 6·7	36.7	36.2	30.4
	81%	45∙5	<b>45</b> ∙1	<b>45</b> ∙4	<b>49</b> ∙2	50∙4	39-6	39.7	38.5
Proportional limit %	12%	52.8	49.3	<b>48</b> ∙0	42·8	<b>48</b> ·8	44-1	54-4	53-3
-	44%	64-3	64·4	61.4	<b>64</b> ·7	64·3	63·0	68·5	67·1
	65%	48·2	42.6	43·1	<b>4</b> 2·8	<b>44</b> ·7	<b>4</b> 3·7	<b>4</b> 8·3	47.1
	81%	37.8	36.3	37-2	33·0	34.6	<b>40</b> ∙3	40·9	44.9

		Percentage inclusion							
	r.h.	0	0.01	0.1	0.2	0.5	1	5	10
Ultimate tensile	12%	53.6	49.6	45.4	46.6	47.5	50·2	<b>41</b> .7	36.3
strength $Nm^{-2} \times 10^{6}$	44%	55.5	49.6	43.1	39.6	46.4	<b>48</b> ·4	36.8	31.7
-	65%	55.5	50-1	42.6	38·2	46.8	50·7	<b>48</b> ∙0	39.0
	81%	41·4	41·2	<b>4</b> 7·7	39.9	50∙5	<b>46</b> ∙0	<b>4</b> 9·1	34.8
Toughness $Jm^{-3} \times 10^5$	12%	100	<del>99</del> .7	91·3	67·0	89·3	<b>89</b> ·1	88·9	82·7
-	44%	92.9	88·7	75·4	59·3	77.7	82.8	66·0	58.3
	65%	105.7	99·1	75.5	59.6	80.5	92.6	94.9	81·2
	81%	<b>88</b> ∙3	<b>87</b> ∙2	86.8	60.8	97·3	<del>89</del> .8	101.6	71.1
Young's modulus	12%	124.6	111.7	95.8	108-2	108-2	113-3	94·3	<b>78</b> ∙0
$Nm^{-2} \times 10^{7}$	44%	143·0	124.7	114.7	105.6	128.9	137-5	113.6	105-5
	65%	111.5	106·0	93.9	<b>80</b> ∙7	94.9	104.4	110.5	<b>79</b> ∙8
	81%	<b>89</b> ∙7	100.3	<b>96</b> ∙7	<b>79</b> ∙7	91.9	97·1	101.7	<b>73</b> ∙8
Elastic resilience	12%	3.4	2.7	2.6	3.6	3.2	3.1	2.5	1.7
$Jm^{-3} \times 10^{5}$	44%	4.6	4.7	3.6	3.3	3.6	3.7	2.6	1.9
	65%	3.4	2.5	3.1	2.4	2.7	2.7	2.1	2.2
	81%	1.8	1.6	1.9	2.3	2.6	1.7	2.0	1.0
Elongation at	12%	46.5	46.9	45.6	32·0	<b>43</b> ·0	41.9	<b>50</b> ·1	58.7
fracture %	44%	<b>46</b> ·0	39.0	38.6	35.0	38.7	38.9	<b>41</b> ·5	<b>48</b> .8
	65%	46.7	<b>45</b> ⋅8	<b>40</b> ∙0	35.7	<b>40</b> ∙0	41.9	44·0	50.6
	81%	45.5	<b>4</b> 0∙5	<b>4</b> 0·3	34.9	<b>44</b> ·8	44.5	47.7	52.6
Proportional limit %	12%	52.8	<b>49</b> ·0	<b>49</b> ·3	58.9	53.9	52.1	<b>50</b> ·8	42·0
	44%	64·3	68·9	67.1	66.2	65·2	64.6	65.5	61.5
	65%	48·2	46.2	56-2	50·5	46.8	<b>45</b> ∙6	43·6	45·4
	81%	37.8	<b>43</b> ∙6	40∙6	<b>4</b> 7∙0	<b>42</b> ∙0	37.4	39.9	32.7

Table 2. Mechanical properties of methylcellulose films. Values represent the mean of 10 replicate measurements. Inclusion: PEG 400

Table 3. Mechanical properties of methylcellulose films. Values represent the mean of 10 replicate measurements. Inclusion: PEG 600  $\,$ 

				Per	rcentag	e inclus	ion			
	r.h.	0	0.01	0.1	0.2	0.5	1	5	10	
Ultimate tensile	12%	53.6	<b>41</b> .8	<b>4</b> 8·7	39.6	47·7	<b>48</b> ∙0	<b>41</b> ·3	37.2	
strength Nm <sup>-2</sup> × 10 <sup>6</sup>	44%	55.5	43·0	44·4	38.2	47.1	47·1	37.3	33.8	
-	65%	55.5	43·5	47·1	36.8	45·9	46.3	41·1	39.2	
	81%	41·4	44·3	<b>45</b> ∙4	<b>40</b> ·1	<b>46</b> ∙5	49.9	<b>4</b> 3∙5	39.8	
Toughness $Jm^{-3} \times 10^5$	12%	100	78·9	85.7	41.5	<b>76</b> ⋅8	86.7	<b>75</b> ·8	<b>80</b> ·9	
•	44%	92.9	<b>76</b> ∙8	72.1	53·8	70·9	72·1	68.9	78·9	
	65%	105.7	87·9	81·7	53·8	63·1	76·7	71·0	82.5	
	81%	88·3	74·6	78·4	52·2	<b>79</b> ∙0	<b>88</b> ∙0	<b>82</b> ·7	<b>89</b> ·1	
Young's modulus	12%	124.6	89.3	108.8	106-3	113-3	119.4	104.7	83·5	
$Nm^{-2} \times 10^{7}$	44%	143·0	126.8	129.2	108·7	130.8	138-2	118-4	101-4	
	65%	111.5	91·6	101·6	89-6	105.5	106-5	<del>9</del> 8·7	83·8	
	81%	89·7	<del>99</del> .7	<b>88</b> ·7	85.8	94·1	101.4	<b>98</b> ·3	78.5	
Elastic resilience	12%	3.4	2.0	2.8	2.8	3.0	2.6	2.6	1.9	
$Jm^{-3} \times 10^{5}$	44%	4.6	2.8	3.4	3.1	3.8	3.7	2.6	2.3	
	65%	3.4	2.5	3.0	2.3	2.7	2.6	1.9	1.9	
	81%	1.8	1.7	1.9	1.7	2.4	2.0	1.8	1.6	
Elongation at	12%	46.5	45.5	<b>41</b> .8	22.9	36.8	39.4	41.6	56.9	
fracture %	44%	<b>46</b> ∙0	45-5	39-4	32.7	34.6	33.9	<b>45</b> ∙4	54.6	
	65%	<b>46</b> ·7	<b>45</b> ∙8	42·2	33.6	30.7	37.0	39.0	50·8	
	81%	45.5	38.8	41·0	28·9	37.5	41.5	<b>4</b> 2∙5	58.5	
Proportional limit %	12%	52.8	43.4	48·9	61-1	53.8	50.1	54.9	46.6	
•	44%	64·3	60·0	66.0	66.8	66·1	67.5	65·7	62·0	
	65%	<b>4</b> 8·2	<b>48</b> ∙0	51.6	53.4	50.4	<b>49</b> ·7	46.1	42.5	
	81%	37.8	41.3	38.6	41.1	<b>43</b> ∙8	38.1	41·3	37.4	

Effects due to propylene glycol (Table 4). Propylene glycol had a significant effect on all of the properties, the general trend was for the presence of 0.01% to cause a reduction in the values obtained, the values then increased at 0.1 and 0.2% additive and decreased with further increases in propylene glycol content. The overall effect of the additive

was to weaken the film and to reduce its elongation at fracture. The only exception to the general reduction in values was the proportional limit which showed only small changes in value at different propylene glycol concentrations.

		Percentage inclusion								
	г.h.	0	0.01	0-1	0.2	0.5	1	5	10	
Ultimate tensile	12%	53.6	45·0	42.6	<b>4</b> 4·5	40.0	42·3	30.0	26.7	
strength $Nm^{-2} \times 10^{6}$	44%	55.5	35.6	39.9	44·8	38.9	39.5	32.2	30.3	
	65%	55.5	37.4	46.1	46.4	43.7	40.9	37.2	30.8	
	81%	41.4	41·3	44·4	<b>50</b> ·7	41·8	36.2	34.2	30.5	
Toughness $Jm^{-3} \times 10^5$	12%	100	69·4	69·0	<b>78</b> ·3	67.9	75.6	46.9	<b>40</b> ⋅8	
8	44%	92.9	<b>4</b> 7·5	<b>59</b> .7	69·8	52·2	57.8	<b>4</b> 0·3	32.2	
	65%	105.7	57.4	<b>79</b> ·8	<b>79</b> ∙0	62·7	64·2	46-4	33.7	
	81%	88·3	<b>69</b> ∙2	76·2	92·3	70·4	57.4	<b>48</b> ∙0	36.0	
Young's modulus	12%	124.6	112·2	107.4	105-6	<b>99</b> .5	98·0	73.7	71·2	
$Nm^{-2} \times 10^7$	44%	143.0	105-3	116.5	124.9	109.8	109.8	101.3	102.0	
	65%	111-5	86.6	105-2	101-5	92.5	86.3	83.3	74·9	
	81%	<b>89</b> ·7	87.8	107.8	107.0	95·1	82.8	<b>79</b> ∙0	82.4	
Elastic resilience	12%	3.4	3.2	2.7	2.7	2.3	2.8	1.6	1.3	
$Jm^{-3} \times 10^{5}$	44%	4∙6	3.3	3.4	3.8	3.2	3.1	2.2	2.1	
	65%	3-4	1.9	2.0	2.4	2.4	1.8	1.7	1.6	
	81%	1.8	1.4	1.5	2.1	1.9	1.5	1.4	1.2	
Elongation at	12%	46.5	33.4	36.3	40·9	40.8	42·4	36.7	35-1	
fracture %	44%	<b>46</b> ·0	30.6	34.4	36.5	31.2	35.4	28.4	23.5	
	65%	46.7	34.9	39.6	39.1	33.5	36.3	28·0	24.6	
	81%	45.5	38.6	37.9	42·3	38.4	36.4	30.8	27·0	
Proportional limit %	12%	52.8	57.5	55·2	52·2	52.3	54·3	<b>49</b> ·4	<b>4</b> 7·7	
•	44%	64·3	73·3	69·7	68·1	67.7	65·8	63·7	66.6	
	65%	48·2	46.3	41.6	46.3	46.5	41.9	42·5	47·6	
	81%	37·8	36.0	38.3	<b>39</b> ·7	43·9	42·0	<b>41</b> ·3	43·3	

Table 4. Mechanical properties of methylcellulose films. Values represent the mean of 10 replicate measurements. Inclusion: propylene glycol

Table 5. Results of analysis of variance to test the effects of the inclusions and the relative humidity on the measured mechanical properties of methylcellulose films.

Additive	Mechanical property	Computed F for inclusions	Computed F for r.h.	Computed F for interactions
Lactose	T. strength	35.1	3.2**	5.0
	Young's mod.	16.2	145.4	1.5*
	Toughness	27.2	13.3	4.0
	Elastic res.	18.4	272.8	4.6
	Elong, at fract.	23.0	12.6	2.3
	Prop. limit	24.1	669.5	3.4
PEG 400	T. strength	56.0	8.7	5.5
	Young's mod.	48.1	75.6	6.9
	Toughness	21.5	10.0	2.1
	Elastic res.	66.4	62.4	6.8
	Elong. at fract.	37.5	4.9	1.4*
	Prop. limit	45·2	452·6	5.8
PEG 600	T. strength	34.5	4.5	1.3*
	Young's mod.	36.9	148.5	24.1
	Toughness	17.6	2.2*	1.2*
	Elastic res.	31-2	107.6	4.0
	Elong. at fract.	50.3	0.4*	2.4
	Prop. limit	30.8	679·2	7.8
Propylene glycol	T. strength	70.5	6.7	3.9
	Young's mod.	50·1	62.4	<b>4</b> ·7
	Toughness	64·4	18.2	2.2
	Elastic res.	69.9	228.8	9.8
	Elong. at fract.	23.0	17.9	2.2
	Prop. limit	7.5	1110-3	10.4

\* Not significant at the P=0.05 level. \*\* Not significant at the P=0.01 level.

		Percentage inclusion							
	r.h.	0	0.01	0.1	0.2	0.5	1	5	10
Ultimate tensile	12%	<b>46</b> ·8	27.3	30.4	26.9	28.6	33.7	37.8	30.6
strength Nm <sup><math>-2</math></sup> × 10 <sup>6</sup>	44%	<b>40</b> ·0	29.0	29.4	28·0	37.3	36.4	32.5	27.3
	65% 81%	39.7 40∙5	29-0 31-8	32·5 29·7	26·5 27·9	37-5 36-3	36·3 34·8	34·7 37·3	38-8 34-1
Toughness $Jm^{-3}\!\times 10^5$	12%	36·7	27.1	44·4	38·5	41·0	21.7	11.1	5.5
	44% 65%	25.7	25.9	30.4	25-2	30.7	22.8	11.3	12.7
	81%	28.4	14.8	19.5	18-4	21.0	24.6	20.5	13.1
Young's modulus	12%	135.5	91·4	99·4	89.6	82.1	108.1	142.8	178.8
Nm $^{2} \times 10^{7}$	44% 65%	134.0	125.0	122-1	106.0	13/*/	131.3	126.4	129.3
	81%	127.0	125.5	99.9	102.2	131.0	112.1	127.5	134.3
Elastic resilience	12%	3.6	1.3	1.7	1.3	2.3	2.0	3.7	2.8
$Jm^{-3} \times 10^{3}$	44%	3.5	2.5	2.6	2.5	3.4	3.1	3.1	2.6
	81%	1.4	1.6	1.9	1.8	1.9	1.8	1.8	2.7
Elongation at	12%	15.6	19.9	28.7	27.7	28.6	12.7	5.7	3.5
fracture %	44%	12.8	21.2	20.5	17.5	[4·9 16.3	12.5	6·9 8.7	4.6
	<b>81</b> %	14.0	9.0	13.0	13.8	11.4	11.4	10.8	7.3
Proportional limit %	12%	65.6	54.4	59·2	54.9	66·1	60.6	87·0	100.0
	44%	76.4	86.1	85.9	86.4	80.6	77.1	86.1	92.9
	81%	<u>49</u> ·3	60·8	64·5	82·8 70·7	58·3	56·6	56·5	<sup>63.4</sup> 75.5

Table 6. Mechanical properties of maize starch films. Values represent the mean of 10 replicate measurements. Inclusion: lactose

Table 7. Mec	hanical proper	ties of maize starch	ı films. Values ı	represent the mean	n of 10
replicate mea	surements. Inc	lusion: PEG 400			

		Percentage inclusion							
	r.h.	0	0.01	0.1	0.2	0.5	1	5	10
Ultimate tensile strength $Nm^{-2} \times 10^6$	12% 44% 65% 81%	46·8 40·0 39·7 40·5	44·5 40·9 40·7 41·6	43·8 40·6 42·2 38·3	45·9 46·6 38·8 39·9	44·7 42·3 39·9 39·1	40·8 40·6 40·6 38·5	31.0 33.4 33.6 31.5	*
Toughness $Jm^{-3} \times 10^5$	12% 44% 65% 81%	36·7 25·7 28·1 28·4	35·3 28·8 27·8 32·3	37·4 24·5 27·9 27·4	38·9 36·2 23·6 23·9	24·3 28·3 25·6 25·9	27·4 27·2 27·6 22·9	7·2 9·1 13·2 13·5	
Young's modulus $Nm^{-2} \times 10^{7}$	12% 44% 65% 81%	135·5 134·6 117·7 127·0	122-0 145-7 120-0 118-5	118·8 137·5 130·9 109·9	123·0 144·0 119·6 115·9	133·7 136·4 124·5 124·3	118·0 155·7 126·3 118·4	138-0 150-7 110-8 100-5	
Elastic resilience $Jm^{-3} \times 10^5$	12% 44% 65% 81%	3·6 3·5 2·4 1·7	3.8 3.6 2.8 2.5	3·2 3·7 2·6 2·4	3·5 5·3 2·9 2·4	3·7 4·7 2·9 2·2	3·5 3·4 3·9 2·3	3.6 3.5 3.2 2.6	
Elongation at fracture %	12% 44% 65% 81%	15·6 12·8 14·0 14·0	16·0 14·1 13·5 15·5	17·1 12·1 13·1 14·3	17·0 15·6 12·1 11·9	10·8 13·4 12·7 13·1	13·2 13·3 13·6 11·8	4·6 5·2 7·8 8·5	
Proportional limit %	12% 44% 65% 81%	65·6 76·4 58·7 49·3	67·0 78·5 62·7 56·8	62·1 77·7 59·4 58·1	61·5 83·6 66·7 61·0	69·4 84·0 65·9 57·8	68·8 79·7 76·6 58·7	100·0 95·0 77·7 75·0	

\* Films too brittle to test.

Table 8. Mechanical properties of maize starch films. Values represent the mean of 10 replicate measurements. Inclusion: propylene glycol

		Percentage inclusion							
Ultimate tensile strength Nm <sup>-2</sup> × 10 <sup>6</sup>	r.h. 12% 44% 65% 81%	0 46·8 40·0 39·7 40·5	0.01 43.1 30.2 39.6 38.7	0·1 40·0 27·8 33·8 38·9	0·2 46·6 31·7 32·0 44·0	0·5 48·2 36·9 39·1 46·2	1 31·9 27·3 33·0 33·5	5 32·6 35·7 34·3 34·3	10 29·0 31·0 33·4 30·4
Toughness $Jm^{-3} \times 10^5$	12%	36·7	47·3	50·4	41·2	43·2	28·0	27·2	16-0
	44%	25·7	34·4	32·3	30·4	37·2	30·0	22·4	16-6
	65%	28·1	39·7	29·7	33·5	37·3	23·7	18·0	16-1
	81%	28·4	31·3	36·0	37·4	39·1	29·5	18·6	19-6
Young's modulus Nm <sup>-2</sup> ×10 <sup>7</sup>	12% 44% 65% 81%	135·4 134·6 117·7 127·0	115·1 118·9 131·1 111·8	117·5 110·4 107·5 112·3	131·7 123·5 105·3 124·3	126·5 129·8 137·6 122·6	100·1 106·0 125·1 121·1	106·2 150·9 135·6 119·2	107·2 124·5 128·7 104·3
Elastic resilience $Jm^{-3} \times 10^5$	12%	3·6	3·0	2·3	3·3	4·0	1.8	2·5	1.7
	44%	3·5	2·4	2·1	2·9	3·6	2.3	3·0	2.9
	65%	2·4	3·0	2·4	2·4	3·5	3.4	3·3	2.9
	81%	1·7	1·9	1·9	2·5	3·0	2.8	2·9	2.4
Elongation at fracture %	12%	15·6	21·9	24·9	17·5	17·7	17·6	15·8	10·9
	44%	12·8	21·4	21·7	18·1	19·4	20·2	12·0	10·6
	65%	14·0	19·9	17·4	20·4	18·8	13·9	10·2	9·5
	81%	14·0	16·1	18·4	16·9	16·8	14·2	10·9	12·6
Proportional limit %	12%	65·6	59·1	56·0	62·4	64·9	57·9	63·2	65∙1
	44%	76·4	77·8	77·1	83·2	82·2	80·6	83·1	86∙4
	65%	58·7	67·5	65·8	68·2	78·8	87·3	86·3	80∙5
	81%	49·3	50·9	50·8	55·1	57·3	65·4	74·5	73•0

Table 9. Results of analysis of variance to test the effects of the inclusions and the relative humidity on the measured mechanical properties of maize starch films.

Additive	Mechanical property	Computed F for inclusions	Computed F for r.h.	Computed F for interactions
Lactose	T. strength	23.8	6.5	3.8
2441004	Young's mod	31.8	10.5	10.2
	Toughness	0.10	28.1	15.6
	Elastic res	27.3	31.5	8.6
	Elong at fract	118.7	50.1	19.0
	Prop. limit	148.6	290.2	38.4
PEG 400	T. strength	50.6	12·ż	3.7
	Young's mod.	0.9*	40.8	2.9
	Toughness	65.5	5.7	5.7
	Elastic res.	3.6	73.5	6.3
	Elong at fract.	51.2	1.6*	4.7
	Prop. limit	132.3	251.0	11.4
Propylene	T. strength	50.7	52.9	11.7
glycol	Young's mod.	15.2	13.2	10.5
0,	Toughness	81·4	23.6	4.3
	Elastic res.	3.6	73.5	6.3
	Elong, at fract.	56.5	20.8	2.9
	Prop. limit	73.9	448.5	16.9

\* Not significant at the P=0.05 level. \*\* Not significant at the P=0.01 level.

Inclusions in maize starch films

The results are given in Tables 6-8 with the analysis of variance summarized in Table 9. PEG 600 was not investigated due to the close similarities between its effects and those of PEG 400 in methylcellulose films. Inspection of Table 9 shows that the concentration of additive, the relative humidity of storage and their mutual interactions had significant effects on the results except in two cases. The coefficients of variation for replicate measurements on maize starch films were similar to those for methylcellulose films. With PEG 400 the concentrations of inclusion had no significant effect on Young's modulus and the relative

humidity of storage had no effect on the elongation at fracture.

Effects due to lactose (Table 6). The inclusion of lactose at low concentrations 0.01-0.2% produced a fall in tensile strength and elastic resilience. Above 0.5% of lactose in the film, toughness and elongation at fracture fell whereas Young's modulus rose. Above 5% lactose the elastic resilience and proportional limit both increased. The effect of lactose above 0.5% is therefore to produce a more brittle and weaker film. This may be due to the phenomenon of antiplasticization. Effects due to PEG 400 (Table 7). PEG 400 had little effect on maize starch films at 1% inclusion and less; at 5%, tensile strength and toughness were reduced and the proportional limit was increased. Films containing 10% PEG 400 were too weak to handle and so could not be tested, they were more opaque than films containing lesser concentrations of inclusion and this suggests that PEG 400 is incompatible with maize starch films.

Effects due to propylene glycol (Table 8). As with PEG 400, low concentrations of propylene glycol had little effect on film properties, but above 0.5% tensile strength and toughness fell. Films were obtained with 10% propylene glycol but otherwise the behaviour of films with propylene glycol and with PEG 400 were similar.

There is a problem in finding a mutually soluble material to act as plasticizer for starch films because of its crystallinity. In the case of PEG 400 there was phase separation at 10% inclusion. Although lactose and propylene glycol appeared to be retained within the polymer film it is unlikely that the latter bonded with the polymer matrix. The presence of lactose at higher concentrations caused an increase in Young's modulus and to a lesser extent an increase in tensile strength coupled with a fall in elongation at fracture. These changes are characteristic of antiplasticization and it is conceivable that the polyhydroxylactose could interact with the polyhydroxycellulose of starch to produce this effect.

Although the relative humidity of storage significantly affected the mechanical properties (Table 9) there was no plasticization by the water. This can be related to the nonsolubility of starch films in water. The inclusions tested thus showed only a tendency to weaken maize starch films. How this weakening affects the properties of granules and tablets is the subject of further study.

## **Conclusions**

The presence of the inclusions tested in the methylcellulose

and maize starch films had the general effect of weakening the films. However, with methylcellulose films there were concentrations of inclusion, in the range 0.2-0.5%, at which film properties were altered to a greater extent than at higher and lower concentrations.

Polyethylene glycol 400 and 600 acted as plasticizers for methylcellulose films but plasticization of maize starch films was not seen with any of the inclusions.

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