International Journal of Pharmaceutics, 52 (1989) 213–219
Elsevier

IJP 01767

Determination of the critical stress intensity factor (K_{IC}) of microcrystalline cellulose using radially edge-cracked tablets

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(Received 7 October 1988)
(Accepted 24 November 1988)

Key words: Critical stress intensity factor; Radially edge-cracked tablet; Edge-opening; Diametral compression; Microcrystalline cellulose

Summary

The critical stress intensity factor K_{IC} of microcrystalline cellulose has been determined using two new techniques based on the fracture of radially edge-cracked tablets. Of the two techniques used, edge opening and diametral compression, the former was the one preferred, since it gave the most stable crack propagation and the effects of crack length were minimal. However, the diametral compression test was found to have some value provided that crack lengths were limited to certain ranges. Extrapolation of K_{IC} to zero porosity using an exponential or second-degree polynomial gave values of 2.24 MPa $\cdot$ m$^{1/2}$ and 2.31 MPa $\cdot$ m$^{1/2}$, respectively, for the edge-opening test and 2.98 MPa $\cdot$ m$^{1/2}$ and 2.35 MPa $\cdot$ m$^{1/2}$, respectively, for the diametral compression test.

Introduction

Pharmaceutical materials used in tabletting vary in their deformation behaviour from those that are brittle and consolidate by particle fragmentation to those that are ductile and consolidate by plastic flow. One method of characterising the mechanical behaviour of powders is to determine the resistance to deformation by measuring the change in volume during compaction (Roberts and Rowe, 1985). Although this technique gives an indication of the scale of deformability of materials, it does not give the true mechanical material property describing the brittleness/ductility. A method of determining the brittleness of materials is to determine the fracture toughness R or the critical stress intensity factor, K_{IC}, since both describe the resistance of a material to the propagation of a crack and are related by Eq. 1 (for plane strain),

$$R = \frac{K_{IC}^2(1-\nu^2)}{E} \tag{1}$$

where E is Young's modulus and ν is Poisson's ratio. In a previous paper Roberts and Rowe (1987) introduced the concept of a brittle–ductile transition in pharmaceutical materials and by studying particle size effects were able to calculate the fracture toughness, R, of lactose and a drug (propanolamine derivative). They were able to classify pharmaceutical materials as brittle or semi-brittle – ideal materials for the application of fracture mechanics.

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Recently Mashadi and Newton (1987a) reported the determination of the critical stress intensity factor of microcrystalline cellulose and sorbitol from notched 4-point beam bending. They were therefore able to calculate a fracture toughness, R, of microcrystalline cellulose (Mashadi and Newton, 1987b) using a value for Young's modulus determined using the technique (4-point beam bending) of Church and Kennerley (1983).

All the techniques for the determination of the critical stress intensity factor, K_{IC}, of pharmaceutical materials have involved the preparation of a range of compressed porous beams, requiring special punch and dies and high tonnage presses. The ideal specimen shape for the determination of this factor is the tablet (radial disc), since it is easy to prepare and is the specimen actually used in tablet manufacture. Recently Kendall and Gregory (1987) reported a technique for the determination of the critical intensity factor on such a tablet specimen type by using 3 tests on the radially edge-cracked discs, i.e. (a) edge opening, (b) diametral compression and (c) pin-loading. They concluded that while the first of these was the best test, all the tests had advantages over other common toughness tests because of their exact theory, simple sample preparation, ease of pre-cracking, straight-forward loading and low propagation forces.

In this study the edge-opening disc and the diametral compression tests have been examined using microcrystalline cellulose compacted at various porosities.

Theory

Two conditions are necessary for a crack to grow under static loading, firstly the stress must be high enough to initiate fracture and secondly, the energy released by crack growth must be equal to the energy required to form new surfaces. The stress intensity factor K, is the stress field intensity at the tip of a crack and is a function of the applied load and the test piece geometry, having dimensions of stress × length$^{1/2}$. For tests involving the determination of K, there are 3 modes of crack surface displacement, mode I (opening mode), corresponding to the normal separation of crack walls under the action of tensile stresses. This is the most common crack propagation mode and therefore given the subscript IC. The other two modes are mode II (sliding mode) and mode III (tearing mode).

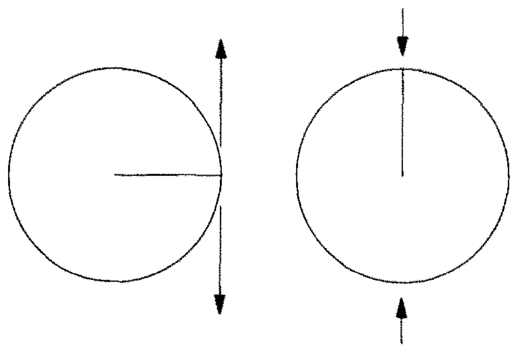

Fig. 1. The two tests used in the fracture of the radially edge-cracked tablets: (a) edge opening and (b) diametral compression.

The two tests considered in this paper the edge-opening and diametral compression tests are shown schematically in Fig. 1. Both these tests involve the introduction of a pre-crack onto the edge of the disc.

Edge opening

The solution for determining K_{IC} for the edge-opening test (Kendall and Gregory, 1987) is given by the equation:

$$K_{IC} = \frac{F}{t}\left[\frac{c}{0.3557(d-c)^{3/2}} + \frac{2}{0.9665(d-c)^{1/2}}\right]\left[\frac{c}{2d}\right]^{-1/2} \tag{2}$$

where F = peak load for cracking, c = the crack length, t = the tablet thickness and d = the tablet diameter.

Compression

The diametral compression test is the most convenient experimental loading technique: in this the cracked disc is loaded between two flat platens. In the absence of a crack, the compressive force F produces a uniform tensile stress across the centre of the loaded diameter and the tensile strength, σ_0 of the disc can be determined as given by the equation.

$$\sigma_0 = \frac{2F}{\pi dt} \tag{3}$$

The use of the diametral or Brazilian test which was originally devised for measuring concrete (Carneiro and Barcellos, 1953) has been used extensively in the pharmaceutical industry for measuring tablet tensile strengths and was originally introduced by Fell and Newton (1968). When a crack is present, it has been shown (Kendall and Gregory, 1987) that this diametral loading is equivalent to a uniform pressure, $2F/\pi dt$, opening the crack faces together with two crack closing forces, $F/\pi t$, applied at the mouth of the crack. If it is assumed that the lateral forces at the crack mouth cancel out, then cracking is driven by only the uniform pressure term and can be described by the equation (Kendall and Gregory, 1987).

$$K_{IC} = \frac{F}{dt}\left[\frac{\pi}{2c}\right]^{-1/2} \frac{1.586}{[1-(c/d)]^{3/2}} \tag{4}$$

Materials and Methods

Avicel PH101 with an initial moisture content of 5.2% was compressed on an F3 tablet compressor (Manesty Machines, Liverpool) using 15 mm diameter flat-faced punches to give a range of porosities. To initiate a radial edge crack, a scalpel blade held in the air jaws of a tensometer (M30K, J.J. Lloyd Instruments, Southampton) was pressed radially on to the edge of the disk using a velocity of 0.1 mm $\cdot$ min^{-1}. Various maximum loads ranging from 30 to 80 N were applied depending on the porosity of the compact. As the peak load was reached the crack extended into the tablet. When the crack had been driven for about 5 mm (this took approximately 1 min), the blade was reversed to arrest the propagating crack. The tablets were removed from the rig and dusted with carbon black to reveal the crack. Crack lengths were measured by a travelling microscope to an accuracy of ± 0.05 mm.

During both the edge-opening and diametral compression tests the forces and displacements were continuously monitored by using an IBM PC-XT connected to the TK30 module (RS232 communications) of the tensometer.

For the edge-opening test, the cracked tablets were gripped in the air jaws and pulled apart using a velocity of 0.2 mm $\cdot$ min^{-1} and the peak load measured. A typical force/displacement curve of the cracking process is shown in Fig. 2. This

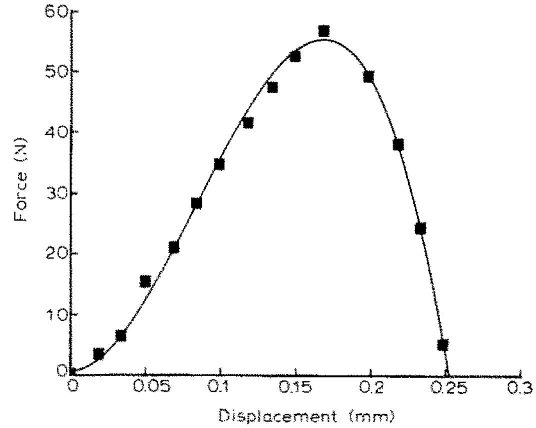

Fig. 2. The force/displacement occurring during the fracture of a tablet for the edge-opening test.

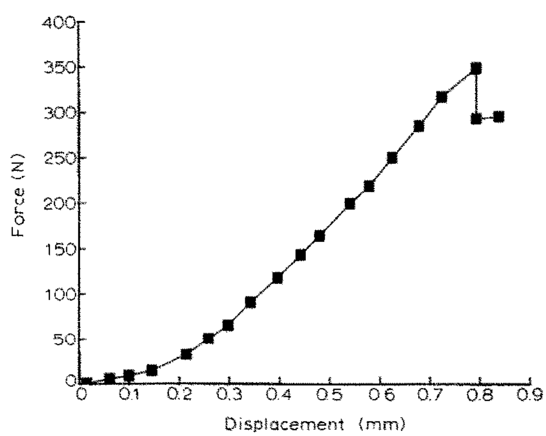

Fig. 3. The force/displacement occurring during the fracture of a tablet for the diametral compression test.

type of curve is typical of that produced when stable cracks are propagated.

In the diametral compression test, the cracked tablets were placed between the platens of the tensometer (with the crack vertical and touching the upper platen) and stressed using a velocity of 0.2 mm · min^{-1}. In this test the resultant force/displacement curve indicated that the cracks were unstable (Fig. 3).

For both tests the effect of different pre-crack lengths was examined for all tablet porosities and only the values from those specimens that gave straight cracks were used in the analysis of K_{IC}.

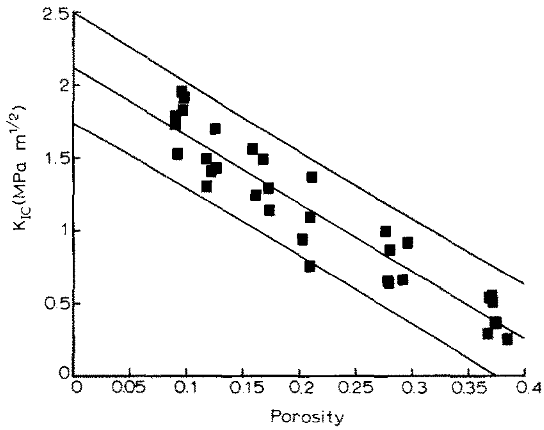

Fig. 5. The critical stress intensity factor, K_{IC}, as a function of tablet porosity using the diametral compression test, with the 95% confidence limits.

Results and Discussion

The effect of crack length

The results from both techniques are shown in a plot of K_{IC} versus porosity (Figs. 4, 5). For the edge-opening technique, crack lengths do seem to have a slight effect on the value of the calculated K_{IC} since comparisons at a porosity of 8.5% indicate that K_{IC} varies from 1.66 to 1.87 MPa · $m^{1/2}$ for crack lengths varying from 4.45 to 7.75 mm. This may be due to inaccuracies in the estimation of crack lengths, since for porous specimens it is difficult to identify the crack tip.

For the diametral compression test the effect of crack lengths was more closely monitored since

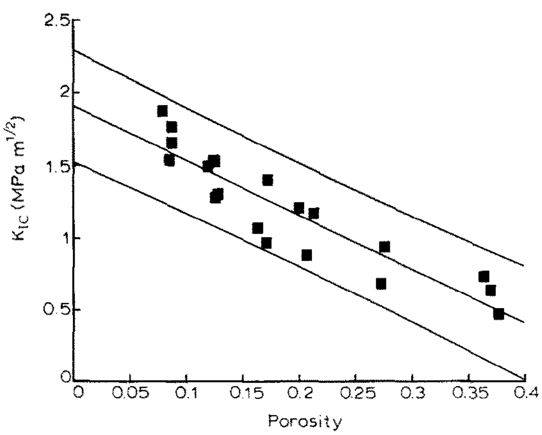

Fig. 4. The critical stress intensity factor, K_{IC}, as a function of tablet porosity using the edge-opening test, with the 95% confidence limits.

Kendall and Gregory (1987) have shown that for polymethylmethacrylate two discrepancies between theory and experiment were evident at crack lengths outside the range of c/d values of 0.34–0.6. Firstly, crack propagation forces were significantly higher which would result in an overestimation of K_{IC}, due to the large plastic regions near to the loading points. Secondly, when the length of the initial crack had reached about half-way across the disc, the cracking force levelled off and the crack became unstable, accelerating rapidly. These two effects were alleviated at least in part by lengthening the initial pre-crack and applying the load over a reduced area using 1.5 mm thick steel fingers. However, the large plastic zones were still evident and the only improvement was better crack stability. It was therefore concluded that the theory required substantial modification to take account of these plastic zones.

In Fig. 6 the effect of crack length on measured cracking force F, at various porosities is highlighted. This factor causes a huge variation in the calculated K_{IC} values, e.g. at a porosity of 16.9%, K_{IC} varies from 0.661 MPa · $m^{1/2}$ to 2.384 MPa · $m^{1/2}$ for the crack lengths varying from 2.75 to 9.55 mm. Although there is a levelling of the cracking force at crack lengths greater than 9 mm (this agrees with that shown by Kendall and Gregory, 1987), there does not seem to be the high propagation forces associated with plastic zones

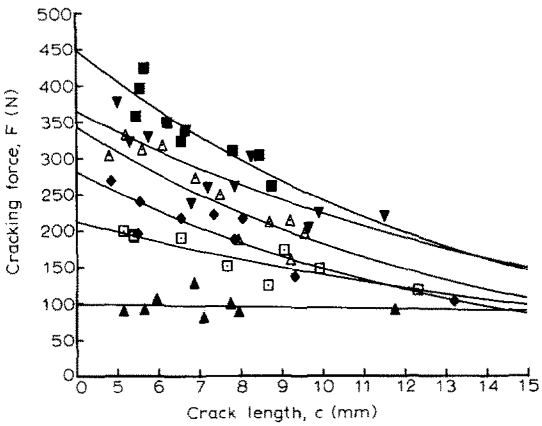

Fig. 6. The effect of crack length on the cracking force using the diametral compression test for tablet porosities of 9.5% (■), 12.0% (▼), 16.9% (△), 20.4% (◆), 28.0% (□), and 37.2% (▲)

that were apparent with polymethylmethacrylate at $c/d \leq 0.34$. However, there was some evidence of squashing of the tablets at the contact with the platens. Hence for the diametral compression test it was decided to limit the results to those that had crack lengths between c/d values of 0.34–0.6.

Comparison of data

The results from the edge-opening and diametral compression tests are shown in Fig. 4 and 5. Using a linear extrapolation, values of K_{IC} at zero porosity were found to be 1.91 $MPa \cdot m^{1/2}$ and 2.12 $MPa \cdot m^{1/2}$, for the edge-opening and diametral compression tests, respectively. These compare with a value of 1.21 $MPa \cdot m^{1/2}$, from 4-point beam bending (Mashadi and Newton, 1987b). Although the values do not compare exactly, there is reasonable agreement between both groups. The lack of exact agreement may be due to a number of factors. Firstly, in the 4-point beam technique used by Mashadi and Newton (1987b), the loading rate was 0.025 $mm \cdot min^{-1}$ whereas for this experiment the rate was 0.2 $mm \cdot min^{-1}$. At higher loading rates the value of the fracture stress would be expected to increase and therefore K_{IC} will be greater than at lower rates. However, the 10-fold increase in rate between the two test procedures would not be expected to increase the critical stress intensity factor by such an amount since it has been shown that for sodium chloride (Roberts et al., 1989) K_{IC} increases from about 0.18 to 0.22 $MPa \cdot m^{1/2}$ for a change in loading rate from 1.98 to 6000 $mm \cdot min^{-1}$ (an increase of 0.04 $MPa \cdot m^{1/2}$ for a 3000-fold increase in loading rate). Therefore it is likely that loading rate differences can be discounted in this study.

Secondly, the moisture content of the beams from both workers may not be the same and this may account for the differences. It is well known for instance, that water can interact with the crack tip in a number of ways depending upon the material, e.g. for soda-lime glass K_{IC} was found to decrease with increasing amounts of water vapour (Wiederhorn, 1967) while for wood fractured along the grain water vapour caused an increase in the fracture toughness R (Jeronimidis, 1979).

Finally crack sharpness effects may account for the differences between the 4-point beam and the radial disc tests, since in the former test it is difficult to insert sharp cracks (Freimen, 1983). This aspect of crack sharpness in the 4-point beam test has been studied by Simpson et al. (1974) who showed that K_{IC} was dependent on the sawn notch radius and became constant below a critical notch radius. It has therefore been suggested by Munz et al. (1980) that the preferred crack geometry for the 4-point beam test is the chevron

TABLE 1

The critical stress intensity factor, K_{IC} of microcrystalline cellulose using various extrapolations to zero porosity

Technique	Equation	K_{IC} at/zero porosity ($MPa \cdot m^{1/2}$)	Parameters	S.E.
Edge-opening	LINEAR	1.911	$a = -3.760$	0.552
Compression	LINEAR	2.119	$a = -4.673$	0.716
Edge-opening	(5)	2.237	$b = -3.626$	0.139
Compression	(5)	2.979	$b = -5.203$	0.204
Edge-opening	(6)	2.307	$f_1 = -8.159$, $f_2 = 9.711$	0.147
Compression	(6)	2.350	$f_1 = -7.117$, $f_2 = 5.205$	0.169
Edge-opening	(7)	2.470	$b = -4.726$, $c = 2.430$	0.141
Compression	(7)	2.035	$b = -1.169$, $c = -8.594$	0.195

notch, which produces a sharp stable crack, with the added advantage that K_{IC} is determined from the maximum load without the need for pre-crack length measurements.

Extrapolation of K_{IC} to zero porosity

It may be expected that the relationship between K_{IC} and porosity will be of an exponential type since it has been shown for a number of mechanical properties that this is the case, e.g. fracture toughness (Rice et al., 1978), modulus (Spriggs, 1961), tensile strength (Ryshkewitch and Duckworth, 1953), and indentation hardness (Leuenberger, 1982). In Table 1 the regression analysis of the results from both tests are examined by comparing different equations relating K_{IC} to porosity, P.

$$K_{IC} = K_{IC_0} \exp(-bP) \tag{5}$$

$$K_{IC} = K_{IC_0}(1 - f_1 P + f_2 P^2) \tag{6}$$

$$K_{IC} = K_{IC_0} \exp[-(bP + cP^2)] \tag{7}$$

These relationships between a mechanical property and porosity have been examined extensively by Phani and Niyogi (1987) for modulus and therefore it seems logical to apply these types of equations to our results. For the two methods of testing, the polynomial type relationship, Eq. 6 seems to give the lowest standard errors (S.E.) and give K_{IC_0} values which are closest in value. This is not surprising, since this type of equation can be used to fit most experimental results. The equation which gives the worst fit is the linear plot and this is as expected since the experimental points clearly lie on a curve (Fig. 4, 5). The two related equations, Eqns. 5 and 7 give similar standard errors. However, for the edge-opening test Eq. 5 gives the lowest standard error of all but gives the biggest differences in the extrapolated K_{IC} values. In analysing the porosity dependence of fracture energy R, Rice et al. (1978) recognised that firstly, R should be proportional to the fraction of solid area fractured, and secondly, fracture will follow the path of maximum pore and minimum solid cross-sectional area. They analysed an exponential type equation and calculated the fracture energy R as a function of porosity for a variety of pore shapes and thus gave a range of b values characteristic of that pore shape, since R and K_{IC} are linked by Eq. 1. A similar relationship between K_{IC} and porosity and pore shape would be expected and therefore it is suggested that this type of equation may be of use in gaining a better understanding of the fracture process in porous materials. However, further comparisons using a variety of materials will need to be analyzed before one equation can be used universally.

Conclusion

The edge-opening test is the preferred one of the two tests, since the effect of crack length is minimal and the results on microcrystalline cellulose are reasonable when compared against other techniques. The diametral compression test can still be used provided that crack lengths are limited to c/d values of 0.3–0.6. However, great care should be taken in the interpretation of the results since for more plastic materials, the presence of the plastic zones can affect the determination of K_{IC} to a great extent (Kendall and Gregory, 1987). Furthermore, it is suggested that the linear type extrapolation of K_{IC} to zero porosity should not be used, since it gives an underestimation of K_{IC} and so either an exponential or the second-degree polynomial equation should be used.

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