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Axial tensile fracture of microcrystalline cellulose compacts

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Abstract

An adapted tensile stress methodology for the fracture of microcrystalline cellulose (MCC) tablets has been investigated and implemented. The application of the generally applied linear elastic fracture mechanic (LEFM) parameters used to describe the fracture behaviour of these porous systems has been discussed. The application of an effective crack length concept, comprising of the notch depth and a process zone length designated Δc, has enabled the localised non-linear response of the MCC tablets to be characterised in a quantified manner. The requirement of the composite value Δc is postulated to be a direct result of the internal properties of the tablet formed during the compaction process due to its strong dependence on porosity. The high compact relative density creates a greater possibility for both local small-scale plastic yielding at the crack tip, commonly found in polymer materials and microcracking ahead of the crack tip, typically observed in the fracture of ceramics. The extrapolated value of K_{IC0} of 0.72 MPa $m^{0.5}$ found in this work lies within the range found in literature for this material indicating that the adopted procedure is acceptable for the determination of the resistance to fracture of MCC compacts.

Keywords: Microcrystalline cellulose; Fracture mechanics; Effective crack length; Tensile strength

1. Introduction

The manufacture of tablets of powder compacts of MCC within the pharmaceutical industry is of key importance as it provides the commonly used method of active ingredient dosage: oral ingestion. The characterisation of powder compacts through mechanical testing provides crucial information for the development of the compaction and processing stages of tablet production. The manufacture of multilayer tablets within the pharmaceutical tabletting industry has been available for at least the last 50 years (Cleaver, 1969; Little and Mitchell, 1949) with increasingly more complex designs and formulations being created. The sequential compaction of powder layers has enabled tailored release profiles of the active ingredient to be obtained (Abdul and Poddar, 2004). The multilayer formulation has however also created new challenges for the mechanical strength testing methodologies. Bilayer tablets have shown to catastrophically delaminate at the non-planer interface between the adjacent compacted layers (Inman et al., 2007). The fracture process that occurs is thought to be caused by internal tensile stresses normal to the plane of fracture as found during the fracture of single compacted tablets during capping (Nystrom et al., 1978). The experimental quantitative strength determining fracture process of the interface of bilayered tablets has therefore created a demand for a mechanical fracture methodology where a tensile stress is developed orthogonal to the interface between adjacent compacted layers, along the axial plane. Commonly applied methodologies for the determination of the relative strength or resistance to fracture of materials include indentation (Miyazaki et al., 2007), three- and four-point beam bending (Mashadi and Newton, 1987; York et al., 1990; Roberts et al., 1993; Hancock et al., 2000), deeply double edge notched tension (Gong et al., 2005), double torsion (Mashadi and Newton, 1988), radially edge cracked disks (Roberts and Rowe, 1989; Kendall and Gregory, 1987), diametrical compression (Mohammed et al., 2005; Rudnick et al., 1963) and friability testing (Shepler and Whitney, 1978; Riippi et al., 1998; Schultz and Kleinebudde, 1994). These fracture methods, however, either require the production of specimens in unique geometries which are not found within industrial processing, or do not result in an applied tensile stress acting in the axial plane of a cylindrical compact. A

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fracture methodology and associated analysis that seems to have been overlooked in recent times is the axial tensile test developed by Nystrom et al. (1977). During testing a tensile stress develops which is normal to the axis of the tablet and hence has shown a good correlation with the observed capping tendency of different pharmaceutical materials (Nystrom et al., 1978). Karehill et al. (1990) have shown that the fracture of bilayered pharmaceutical tablets can be achieved using this methodology when investigating the effects of surface roughness on adhesion. However no substantial investigation into the mechanics of fracture was undertaken.

2. Fracture mechanics

The theoretical study and application of the mechanics of crack growth within pharmaceutical materials is important as it helps to more fully characterise the failure process of these tablets and may elucidate possible solutions to prevent tablet cohesive failure within future manufacturing. The porosity of powder compacts makes the prediction and measurement of tablet strength relatively complex: inherent space within the compact creates the possibility of strength weakening internal flaws or cracks. Due to crack propagation the bonded particles separate sequentially (Kendall, 1988), meaning the application of a Griffith (Griffith, 1920) style energy balance is appropriate. Griffith theory, the basis for linear elastic fracture mechanics (LEFM), assumes that the deformation at a crack tip within a material is completely elastic and thus no plastic deformation of the material occurs. The experimental determination of LEFM parameters used to describe the material properties of MCC during fracture has been published by various authors (Mashadi and Newton, 1987, 1988; Roberts and Rowe, 1989; York et al., 1990; Roberts et al., 1993; Hancock et al., 2000). Within these publications there has been no attempt to introduce the application of non-linear fracture mechanic principles to account for the small zone (ca. 1 mm, for example) of plastic deformation ahead of the crack tip, which is surprising given the inherent ductile nature of these materials. MCC compacts are consolidated particulate solids made of cellulose polymers and therefore it would seem logical that the fracture behaviour of MCC tablets is a combination of the characteristic behaviour of both ceramic fracture (brittle and unstable) and polymer fracture (displaying relatively small scale local plastic yielding). Thus the LEFM assumption of a fully elastic response at a sharp crack tip appears to be an erroneous simplification of a more complex fracture propagation process. Small scale plastic yielding at a crack tip does not automatically mean that LEFM cannot be applied, merely a 'modified', as opposed to an apparent, crack length must be included in the fracture parameter calculations (Irwin, 1957, 1961) to fully understand the material response of MCC compacts to a propagating crack. The commonly quoted LEFM parameter K_C or critical stress intensity factor provides an indication of the brittleness of a material by evaluating the stress field near a crack tip. As discontinuities such as pores or flaws, of a variety of sizes and shapes, will exist in the material concentrating the local stresses, K_{IC} is usually obtained from the fracture stress of a sample containing a significantly dominant pre-notch or crack of known length (Eq. (1)).

$$K_C = Y\sigma c^{0.5} \tag{1}$$

where Y is a polynomial function based on the geometry of the sample and loading, σ the fracture stress and c is the crack length. When relatively small scale crack yielding is present, c is arbitrarily replaced by an effective crack length $\hat{c}$ (Eq. (2)) where the crack length has been extended by the radius of a supposed process or inelastic zone Δc.

$$\hat{c} = c + \Delta c \tag{2}$$

The exact interpretation of the process zone is fairly ambiguous. However, it is well understood that at least for ductile materials the process zone is a key parameter that prescribes the toughness (Adams et al., 1989) and therefore seems to be a logical inclusion for the fracture parameter calculations for MCC. The use of LEFM to characterise the fracture behaviour without this process zone extension merely provides a quantitative 'estimate' of the fracture mechanism, suitable for general comparison and the ranking of materials. The main purpose of this current work is to develop the analysis of the MCC compacts material response to fracture utilising an axial tensile stress methodology. By computationally including any process zone, which may be present at the crack tip it is hoped a more rigorous analytical approach to the fracture of these porous bodies will improve the understanding of the mechanical response to fracture of MCC compacts.

3. Experimental procedure

3.1. Materials

Microcrystalline cellulose (MCC) was used as supplied in the form of Avicel PH102, FMC Corp., Philadelphia, USA. The material has a mean particle size 95.3 μm averaged over three runs using a HORIBA LA-950 Wet ver. 3.2 particle seizer (Particle Technology Ltd., Derbyshire, UK). The samples were dispersed in deionised water and no wetting agent or surfactant was used. No ultrasonics was applied to provide the closest possible representation of the ambient dry powder properties. Preliminary runs of the sample initially after wetting and after a period of 5 min showed that no swelling of the particles occurred. MCC has a true (pycnometric) density of 1577 kg/m^3 (obtained from the average of 10 runs using an AccuPycnometer 1330, Micromeritics, Bedfordshire, UK). MCC is a largely ductile material and hence deforms in an extensive plastic manner under a compressive stress (Roberts and Rowe, 1987).

3.2. Compaction

An unlubricated flat-faced punch and die set was utilised, which was manufactured from optically polished hardened stainless steel supplied by Specac Ltd., Kent, UK. The diameter of the die was 20 mm producing tablets of 20 mm diameter compressed to a maximum pressure of between 12.7–156 MPa. The weight of the samples was varied to provide a constant aspect ratio of the tablets of 0.5 (final compact height of 10 mm). Variability within

the compaction process is likely to be caused by inconsistencies produced during die filling. Attempts to reduce this influence included initial manual vibration of the die and flattening with the upper punch before the compression process proceeded. The tablets were compacted using an EZ-50 Materials Testing Machine, manufactured by Lloyd Instruments Ltd., Hampshire, UK. It was a bench mounted, twin leadscrew machine, which could apply uniaxial forces up to 50 kN using an interchangeable load cell. The machine was supplied with a cross-head guidance system to prevent side loading of the samples under compression. A high resolution location encoder enabled the establishment of the upper punch displacement measurements with an accuracy of 10 μm. A constant upper punch velocity of 0.083 mm/s was specified with an accuracy of 1.6 μm/s. The system was connected to a microprocessor system and operated from a multi-position control console. The software programme NEXYGEN (Lloyd Instruments, Hampshire, UK) was used for data collection via the integral RS232 interface. After compaction the lower punch was removed and the tablet was ejected by the application of a force to the upper punch driving the tablet out of the die. The density of the tablet was then calculated from the measured mass of the sample and the volume calculated from an average of three height measurements taken across the diameter using a micrometer. The experiments were conducted in a laboratory where the ambient temperature ranged from 17.5 to 25.2 °C with a relative humidity range of 31% to 48%.

3.3. Fracture

A modified uniaxial tensile tester was developed and constructed based upon an original design by Nystrom et al. (1977). These tests were performed on the EZ-50 testing machine (as previously described). As shown in Fig. 1a, the tablet was glued to two metal adapters using a cyanoacrylate adhesive (Cyanolit 203, Eurobond Adhesives Ltd., Kent, UK) and left overnight to ensure a good adhesion. The lower adapter was connected to the base of the machine and the upper adapter to an upper moveable load cell via a metal chain: used in an attempt to provide full triaxial movement to facilitate the tablet fracturing by an applied tensile stress normal to the radial plane (predominantly mode I failure). After positioning in the tensile tester a constant displacement of the upper adapter was employed (8.3×10^{-3} mm/s) and the resulting tensile force was recorded. The imposed displacement was continued until the tablet catastrophically fractured. To account for the compliance of the equipment the two metal adapters were adhered to each other with no tablet being present. Using the aforementioned arrangement a tensile stress was then imposed up to the value of the highest fracture force obtained from the MCC compacts. The force/displacement curve was then subtracted from the measured force/displacement curves obtained from the notched samples. Sample preparation of the tablets involved machining with a lathe a sharp 'V' shaped pre-notch, in a similar manner to that of York et al. (1990), into the tablets at the midpoint of the axial length, as shown in Fig. 1b. To determine the applicability of LEFM to MCC fracture initial crack lengths ranging from 0.25 to 4 mm ($c/b < 0.4$) were introduced into the tablets and the measurements with each sample was repeated three times. The imposed crack length range was chosen to meet the criteria for plain strain conditions (Tada et al., 2000). The artificially machined crack exceeded multiple (3–40) mean particle diameters so that during fracture an equilibrium stress condition can be readily achieved (Hancock et al., 2000). The sharpness of the crack is an important parameter during fracture but it could not be accurately specified in the current work, however the cutting tool was sharpened before use.

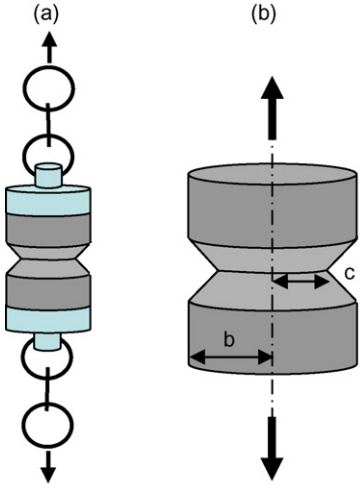

Fig. 1. (a) Schematic of axial tensile stress equipment. (b) Notched tablet.

4. Results and discussion

4.1. LEFM application

A typical reaction force/imposed displacement curve is shown in Fig. 2. The initial non-linearity, up to ca. 0.2 mm, can be ignored as it is due to the alignment of the metal chain between the adapters and load cell. It can be seen that the material displays a linear increase in measured force with respect to a constant increase in the imposed displacement. All the tablets displayed unstable brittle crack propagation: once the

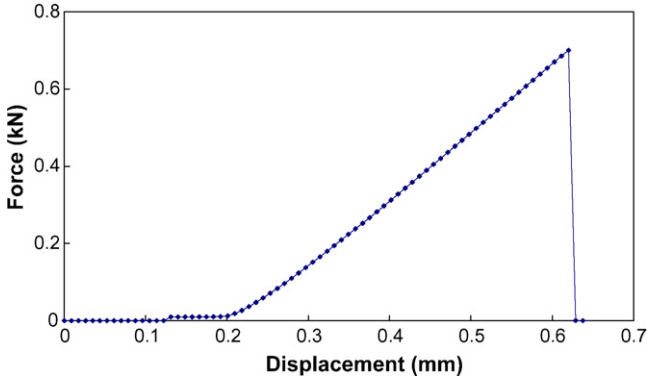

Fig. 2. Typical force displacement curve obtained during the fracture of the MCC compacts showing linear unstable brittle fracture (fracture of tablet compressed to 156 MPa, crack length 0.25 mm).

critical fracture force or energy was reached, the crack propagated instantaneously resulting in the rapid catastrophic failure of the tablets. An infinitely sharp crack can only be achieved via initial loading of the sample to form a 'pre-crack' prior to final failure. As this is unachievable with materials that fail by unstable brittle fracture the introduced sharp 'V' shaped notch appears to provide the most appropriate alternative. The mode I critical stress intensity factor K_{IC} for all the tablets tested was calculated using Eqs. (3) and (4) (Tada et al., 2000). These equations have been initially developed for continuous media subjected to tensile stress using the same geometric specification applied in this work. The fundamental derivation, however, should also be applicable to porous media.

$$K_{IC} = \sigma\sqrt{\pi c} F\left\{\frac{c}{b}\right\} \tag{3}$$

$$F\left\{\frac{c}{b}\right\} = \frac{1}{(1-c/b)^{3/2}} \times \left\{1.122 - 1.302\frac{c}{b} + 0.988\left(\frac{c}{b}\right)^2 - 0.308\left(\frac{c}{b}\right)^3\right\} \tag{4}$$

where σ is the applied stress at fracture, c the corresponding crack length and b is the radius of the tablet (as shown in Fig. 1b). The measured critical energy release rate, G_{CM}, was calculated using the fracture energy (the area under the load displacement curve) obtained during loading (Eq. (5)) and was compared to the calculated value, G_{CC}, obtained from the relationship with K_{IC} for plane strain conditions (Eq. (6)).

$$G_{CM} = \frac{U_F}{\pi(b-c)^2} \tag{5}$$

$$G_{CC} = \frac{K_{IC}^2(1-\nu^2)}{E} \tag{6}$$

where U_F is the fracture energy, b the tablet radius, c the crack length, ν the Poisson's ratio and E is the Young's modulus. The values of both the Young's modulus (E) and Poisson's ratio (ν) vary with the porosity and were obtained using the simplified assumption of linearity and isotropy (Sinka et al., 2003), which is in good agreement with other literatures (Hancock et al., 2000; Michrafy et al., 2004). Initial plots of K_{IC} against c/b for each of the porosities studied clearly showed some variation of the measured facture mechanic parameter with the imposed crack length, a result, which is in contradiction with traditional LEFM theory. A simple analysis of variance was carried out on the repeated estimates of the K_{IC} values for each value of c/b obtained in this work. The commonly quoted 'F ratios' and probabilities of the determined result given the null hypothesis (that a variation in the crack length does not influence the estimated K_{IC} value) for each data set are given in Table 1. It can be seen for all sets of data, with the exception of one with a maximum compaction stress of 28.6 MPa show that there is a statistically significant variation in the calculated K_{IC} value with c/b, thus implying the presence of a process zone ahead of the crack tip. Fig. 3 illustrates this phenomenon showing a logarithmic fit of the data for MCC samples with a porosity of 0.53. A

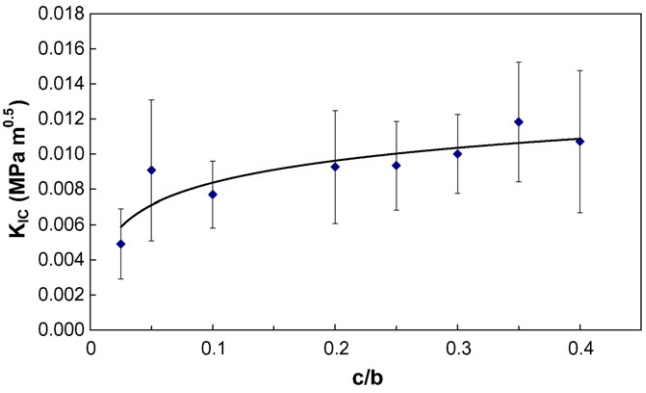

Fig. 3. The non-linear plot of K_{IC} vs. c/b for MCC samples with a porosity of 0.53. The error bars represent one standard deviation. Data fitted to a log curve.

similar trend was observed for the values of the measured critical energy release rate, G_{CM} and the calculated energy release rate, G_{CC}. The application of fracture mechanics has always been plagued with the problem of random inconsistencies due to the presence of variable internal flaws present along the crack propagation path, which is accentuated by the application to these porous systems. This result is no different with the average K_{IC} values having relatively large associated inconsistencies. With the inherent problem of variation of tablet strength within the data sets, as a result of the variation of flow dimensions and strength, it is relatively surprising that the following reported correlations could be achieved. Following the iteration procedure on the K_{IC} values described fully by Adams et al. (1989), an 'Irwin Type' (Irwin, 1961) notional crack length was introduced into all the sets of tablets to account for the apparent non-linearity in the measured K_{IC} against apparent crack length data. The final value of K_{IC} is obtained from the gradient of the linear best fit of the data at the final iteration, taken once the effective crack length remains constant. Fig. 4 shows the results of the first and final iterations for the data set with a porosity of 0.26. A summary of the results computated after n iterations for all porosities studied is shown in Table 2. It can be seen that the measured values of G_C are consistently higher than the calculated values, especially for the more porous and weaker tablets. The experimental variation involved with these

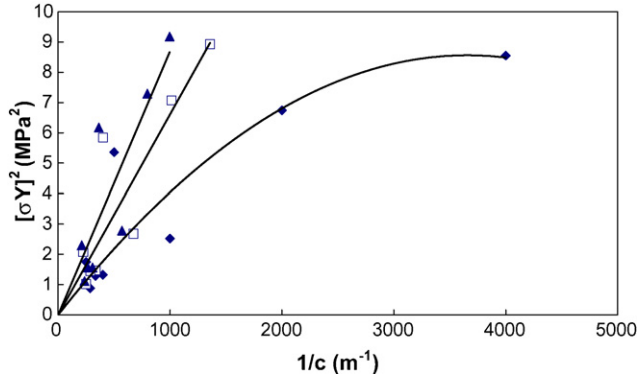

Fig. 4. Iteration results showing increasing linearity for the data set of MCC compacts with a porosity of 0.26. Plot of $(\sigma Y)^2$, from Eq. (1), as a function of $1/c^{(0)}$ (♦), $1/c^{(1)}$ (□) and $1/c^{(n)}$ (▲).

Table 1
Analysis of variance for the K_{IC} estimates with varying c/b values

Compaction stress (MPa)	F ratio	Probability of result given null hypothesis (accurate to 3 d.p.)
12.7	5.32	0.003
28.6	0.78	0.613
39.8	3.12	0.028
50.9	6.16	0.001
79.6	4.54	0.006
95.5	5.10	0.003
127.3	17.78	0.000
156.0	5.17	0.003

experiments is relatively large due to the intrinsic nature of the fracture and the inherent random flaws that are naturally present in such porous systems. This makes the precise measurement of low fracture energy values problematic. The data set compacted to 79.6 MPa produced unexpectedly high values of both K_{IC} and G_C. The reason for this is at present unknown, however, during manufacture and fracture of this particular data set the temperature rose from the ambient temperature range of 23–25 to 31–32 °C. The thermal increase will facilitate the deformation of MCC by plastic flow thus increasing the amount of irreversible compaction energy supplied to the system, albeit not to the extreme reported here. As a consequence the derived effective crack length may also have been slightly over estimated meaning these results should be considered with caution. It appears that the use of an effective crack length concept and value in the K_{IC} calculations justifies the application of LEFM. The physical meaning of the additional crack length or process zone, Δc, is still a matter of some conjecture and is clearly dependant upon the material properties. Common investigations into fracture characterisation involve the positioning of optical sensors to monitor the crack propagation. In this case, however, the geometry of the specimen and speed of the crack propagation makes direct observation impractical. Metallic materials often display a process zone around the crack tip during fracture where ductile yielding occurs. This causes a rise in the fracture force due to the increase in energy required to cause local complex deformation of the material before failure. This interpretation of Δc could be applied to MCC as the material deforms rather plastically under the applied stress. A process zone size of 9.4 μm was reported for a glass particle (mean particle diameter 11 μm) reinforced epoxy resin (Tjernlund et al., 2006). This value can be considered ca. nine to ten times the average distance between cross-linked polymer chains, estimated at 1 μm (Tjernlund et al., 2006). The estimated value of Δc reported in this work ranges from five to ten times the particle size as the compaction stress is varied. Concrete and rock are quasibrittle materials that derive their toughness from a subcritical crack growth that precedes catastrophic failure (Anderson, 2005). This explanation for the effective crack length is inadequate for the results obtained in this work due to the non-linearities, which would have been observed while the samples were under tensile stress. Process zone lengths for concrete have been reported using non-linear fracture mechanics to be up to twice the original imposed crack length (Zhang and Wu, 1999). Process zones found in the fracture of ceramic materials are often the result of microcracking or crack branching: tiny cracks are formed within the bulk material ahead of the crack tip which dissipate strain energy from the sample improving its toughness. Microcracking has been observed with fine grain samples where an increase in fracture energy is required to create multiple new surfaces. Crack branching can be on a multi-grain scale and therefore may represent true changes in fracture area (Wu, 1978). Microcracking within sintered ceramics is attributed to uneven stress distributions, which arise during thermal expansion. This could be considered akin to the uneven internal stress distributions that are created by the imposed pressure patterns created during compaction of MCC: often observed indirectly as an inhomogeneous density distribution (Train, 1957). Microcracking has been seen during the fracture of cortical bone where a process zone size of ca. 3 mm was reported (Cox and Yang, 2007). For the samples studied in this work there is a strong dependency of the value of Δc on the porosity, as shown in Fig. 5 and discussed later. The dependence of Δc on the extent of the porosity was however not seen by Adams et al. (1989) who fractured porous samples composed of a mixture of coarse sand particles and a polymeric binder polyvinylpyrrolidone (PVP) using

Table 2
Fracture mechanic parameters for MCC compacts with different porosities

Compaction stress (MPa)	Porosity	E (MPa)	ν	$K_{IC}^{(n)}$ (MPa m$^{0.5}$)	$G_{CM}^{(0)}$ (J m^{-2})	$G_{CM}^{(n)}$ (J m^{-2})	G_{CC} (J m^{-2})	Δc (mm)
12.7	0.53	670	0.05	0.010	2.41	2.41	0.25	0.50
28.6	0.43	883	0.08	0.026	4.21	4.86	0.79	0.52
39.8	0.38	1240	0.10	0.032	3.28	4.02	0.80	0.52
50.9	0.34	1702	0.12	0.041	3.42	3.91	0.96	0.57
79.6	0.26	2730	0.16	0.096	12.06	14.36	3.29	0.61
95.5	0.26	2784	0.16	0.096	4.99	5.93	3.21	0.75
127.3	0.21	3653	0.18	0.121	9.07	12.50	3.89	1.10
156.0	0.19	4054	0.2	0.178	6.51	8.38	7.48	0.94

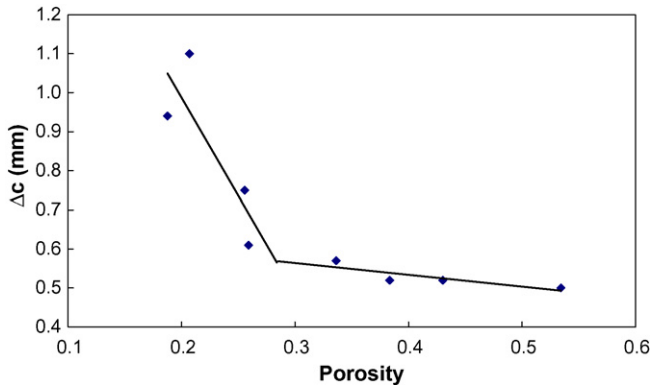

Fig. 5. The two-phase relationship between the process zone length, Δc and the compact porosity. The gradient change at ca. 0.3 corresponds to a compaction stress of ca. 80 MPa, where the structure of individual MCC particles becomes less clear due to increased deformation by plastic flow.

a three-point bending technique. This effect could have been obscured within the seven sets of data reported by Adams et al. (1989) as there were only small variations in porosity and other parameters such as the concentration of PVP and particle size of the sand grains were continuously altered making any direct correlation between effective crack length and porosity difficult to resolve. Examination of the two data sets where only the porosity is varied (A and B) shows the same increase in effective crack length with a decrease in the porosity as was found in the current work. The observed porosity dependence implies that the variation in Δc is a direct result of the intrinsic properties of the tablet, which forms during the compaction procedure. It can be seen in Fig. 5 that there is a two-phase correlation. For porosities above ca. 0.3 the gradient of the correlation is relatively shallow. Once the porosity reaches ca. 0.3 the correlation gradient sharply increases. From Table 3 it can be seen that the applied stress required to reach this critical porosity value corresponds to approximately 80 MPa. This maximum stress value is of the same magnitude of that highlighted by Sixsmith (1977) who determined that the individual particle structure within compacts becomes lost due to particle deformation 'smoothing' the surface at applied stresses of approximately 88 MPa or greater. This infers that the mechanical response of the compact becomes more akin to a solid body than a particulate structure at porosities lower than 0.3. It is therefore inferred that the yielding due to plastic deformation at the crack tip becomes more pronounced at the lower porosities where the individual particles have experienced more deformation by plastic flow.

4.2. Fracture methodology comparison

The critical stress intensity factor at zero porosity, K_{IC0}, obtained by the extrapolation of the measured relationship between the K_{IC} value and the porosity is commonly used to compare fracture data, methodologies and materials. Various interrelationships have been postulated to describe the dependence of the mechanical properties of materials with porosity and a detailed review can be found elsewhere (Phani and Niyogi, 1987). The Spriggs (Spriggs, 1961) exponential relationship was chosen for this work as the trend clearly follows a curve and an exponential has been used by various authors to determine the critical stress intensity value at zero porosity using other mechanical techniques (Roberts and Rowe, 1989; Roberts et al., 1993; York et al., 1990; Mashadi and Newton, 1987, 1988) and has the following form:

$$K_{IC} = K_{IC0} \exp(-AP) \quad (7)$$

where A is a dimensionless constant found to be 8.05 and P is the porosity of the compacted tablet. The correlation, shown in Fig. 6 had an R^2 value of 0.99. The K_{IC0} value of 0.72 MPa $m^{0.5}$ obtained in this work has been compared to published values from literature. It can be seen from Table 3 that there is a wide variation in reported values. The most obvious distinction for the range of K_{IC} values reported is the grade of Avicel and hence, in part, the mean particle size of the MCC investigated. It has been shown that as the initial particle size of the compacted powder decreases the K_{IC0} value of the compact increases due to a decrease in the internal voidage of the compact and hence an increase in the number of contact points available for bonding (York et al., 1990). Avicel PH101 has a smaller mean particle size than Avicel PH102 and thus, according to current wisdom, should have a higher K_{IC0} value. In practice however, a large range of K_{IC0} values has been reported for Avicel PH101 and explanations for the discrepancies between these values have included many variables. Hancock et al. (2000) derived a series of values of K_{IC0} by varying the humidity of the samples. The value discussed here was determined at the RH most consistent with this work. The low value of K_{IC} found by Hancock et al. (2000) was probably due to the sample specimens being relatively small, ~20 mg, meaning any small error in alignment of the testing apparatus could result in uneven stress distributions. The beam and crack dimensions were approaching the size of the micro-structural features of the sample thus by their own admission the compacts tested were around the lower limit for the geometric dimensions applicable for fracture mechanic experiments. K_{IC0} values reported by Mashadi and Newton (1987, 1988) were obtained by fitting their data to a linear relationship meaning that extrapolation to the zero porosity would have resulted in an underestimation of the K_{IC0} value (Roberts et al.,

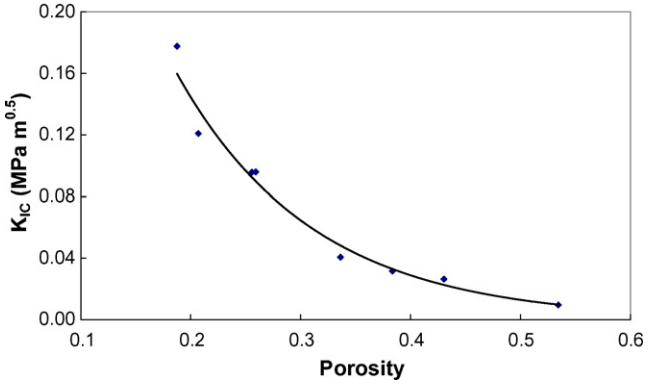

Fig. 6. Porosity–K_{IC} curve for single compaction notched samples of MCC. The K_{IC0} value of 0.72 MPa $m^{0.5}$ is obtained by extrapolation, to zero porosity, of the fitted exponential line (R^2 value of 0.99).

Table 3
Reported values of the extrapolated critical stress intensity factor at zero porosity (K_{IC0}) for MCC

Test	Notch type	K_{IC0} (MPa m$^{0.5}$)	Avicel grade	Reference
Radially edge crack disc (compression)	Pre-crack	2.98	PH101	Roberts and Rowe (1989)
Radially edge cracked disc (tension)	Pre-crack	2.24	PH101	Roberts and Rowe (1989)
Double torsion	Pre-crack	1.81	PH101	Mashadi and Newton (1988)
Four-point SENB	V	1.21	PH101	Mashadi and Newton (1987)
Four-point SENB	Straight	0.87	PH101	York et al. (1990)
Four-point SENB	Straight	0.76	PH102	York et al. (1990)
Three-point SENB	Razor	0.76	PH101	Roberts et al. (1993)
Axial tensile test	V	0.72	PH102	This work
Three-point SENB	Sawn	0.69	PH101	Roberts et al. (1993)
Three-point SENB	V	0.46	PH101	Hancock et al. (2000)

1993). Inaccurate measurement of notch lengths could result in either underestimated or overestimated values of K_{IC} being reported. With MCC tablets being inherently porous the crack tip could actually be positioned at internal void spaces making identification of the actual length difficult (Roberts and Rowe, 1989). Any increase in the loading rates of the samples may affect the sample toughness (York et al., 1990). At higher loading rates the value of K_{IC} would expect to increase (Roberts and Rowe, 1989) due to viscoelastic effects. It has, however, been reported that for a 3000-fold increase in loading rate the K_{IC} value of sodium chloride only increased from 0.18 to 0.22 MPa m$^{0.5}$ (Roberts et al., 1989). Although a slightly more brittle material than MCC this result seems to suggest that loading rate only has a limited effect on fracture toughness compared to other variables. The sharpness of the pre-notch or crack may have a significant effect upon the measured value of K_{IC}. Stressing of a blunt notch (such as a straight or sawn notch used by York et al. (1990) and Roberts et al. (1993) may cause a pre-crack to form before rapid fracture meaning an underestimation of the crack length could occur resulting in an underestimation of the K_{IC0} value (Munz, 1983). The variation in K_{IC} values with the crack length found in this work has also been reported by York et al. (1990). A range in K_{IC} values of 0.42–0.51 MPa m$^{0.5}$ was seen when the crack length was varied between 0.5 and 1.5 mm. Roberts and Rowe (1989) observed a range of K_{IC0} values between 0.66 and 2.38 MPa m$^{0.5}$ during investigations using a radial-edge-cracked disk loaded in compression with cracks in the range of 2.75–9.55 mm. Instead of introducing an Irwin Type modification as used in this work to account for the non-linear LEFM behaviour shown by the material this effect was considered to be the inevitable result of MCC displaying a rising crack growth resistance curve. Application of an effective crack length should account for discrepancies in the notch geometry and sharpness as any energy required to form a pre-crack will be accounted for within the process zone at the crack tip. It would be interesting to apply the iteration procedure used in this work to the data reported in literature to see if more consistency in K_{IC0} values would be obtained, however due to the K_{IC0} being an extrapolated parameter variation in the reported data is likely. Taking into account the large variations in the reported K_{IC0} values for MCC the tensile stress method proposed in this work has provided a more than adequate technique for the assessment of certain fracture parameters. The methodology has thus been applied to bilayer tablets to provide a quantitative method to determine interfacial strength (Inman et al., 2007).

5. Conclusions

An adapted tensile stress methodology has been proposed and utilised for the main purpose of determining a rigorous quantitative analysis of axial fracture of microcrystalline cellulose (MCC) tablets. The proposed methodology develops an axial tensile stress orthogonal to the radial plane within a compact and therefore should provide a simple quantitative technique for the determination of the interfacial strength of complex formulations such as bilayered and multilayered pharmaceutical compacts. Previous utilisation of fracture mechanic techniques with pharmaceutical compact specimens have attempted to determine the definitive mechanical properties of commonly used pharmaceutical ingredients through the application of linear elastic fracture mechanic (LEFM) principles. The applicability of LEFM principles to a MCC compact has been discussed and it has been found that the material does not display an entirely elastic response around the crack tip as dictated by the traditional simple LEFM theory. To account for this observation a more rigorous fracture mechanic analysis has been undertaken. An Irwin Type effective crack length, which includes a process zone around the crack tip, has been included in the corresponding K_{IC} calculations. Due to the inherent ductile nature of the material and the crystalline structure, which is akin to ceramic materials, the process zone is considered to be a combination of plastic deformation at the crack tip and microcracking where tiny cracks are formed ahead of the crack tip. The strong dependence on the compacted size of the process zone with the sample relative density implies that the mechanism of the fracture process is defined by the intrinsic properties of the tablet, which are a direct result of the internal stress distribution caused during the compaction. The complete and correct analysis of the fracture process of MCC tablets has been reported in this work; however, if the data required is merely for the purpose of a simple ranking of materials then the iteration procedure presented here is not essential to the calculation of the fracture mechanic parameters. However, as the procedure is fundamentally correct, relatively simple to implement and may provide a more complete understanding of the fracture process of MCC compacts the utilisation of the Irwin Type process zone extension to

LEFM is advised. The calculated K_{IC0} value of 0.72 MPa $m^{0.5}$ produced in this work has been compared to other fracture procedures and the large range of K_{IC0} values for MCC has been discussed. The tensile stress fracture methodology is considered a suitable technique for the description of the fracture of MCC tablets. Bilayer tablets of MCC have been manufactured to varying compaction stresses and fractured using this tensile stress method (Inman et al., 2007). Further results will be disclosed in a future publication.

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