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Determination of the Critical Stress Intensity Factor (K_{IC}) of Compacted Pharmaceutical Powders by the Double Torsion Method

A. B. MASHADI AND J. M. NEWTON

The School of Pharmacy, University of London, 29–39 Brunswick Square, London WC1N 1AX, UK

Abstract—The critical stress intensity factor K_{IC} has been determined for compacts prepared at different pressures from both Avicel PH101 and sorbitol 'Instant', by means of a double torsion method. This method avoids the need for displacement measurement and the introduction of notch into the specimens, both of which present problems with pharmaceutical materials. Measurements of fracture loads for such specimens were reproducible. The values of K_{IC} derived for both materials were found to increase linearly with increase in compaction pressure and linearly with decrease in specimen porosity. Extrapolation of the latter relationship to zero porosity gave values of 1·81 and 0·69 MN $m^{-3/2}$ for the K_{IC} of Avicel PH101 and sorbitol 'Instant', respectively.

There are a number of industries which utilize the ability of powders to form solid compacts when subjected to compression in a punch and die system. The apparently simple process hides a complexity of stress states within the powder such that, to date, there is no defined theoretical analysis of the process which relates powder properties and process variables to those of the final product. The mechanical properties of the powder have an influence on the stress state induced by the compaction and on the properties of the compact. It is important, however, to realize that powders of a wide range of materials can be compacted, ranging from brittle materials, such as ceramics, to materials which are ductile, such as metals. In terms of placing pharmaceutical materials on the brittle/ductile scale, work is only just beginning; see for example, Stanley & Newton (1977), Roberts & Rowe (1985) and Mashadi & Newton (1987a, b). In general, pharmaceutical materials tend towards the brittle rather than the ductile end of the mechanical spectral range.

This tendency towards brittleness is significant in that the manufacturing process of tableting is dynamic. It is in the defining of the dynamic strength properties of brittle materials and an understanding of their mechanical properties that progress is now being made; see, for example, Curren et al (1987). One of the problems of brittle materials is the absence of a yield value, which provides a useful characterization of ductile materials. The recent approach for characterizing brittle materials is via "fracture mechanics". This is based on the work of Griffith & Irwin (1968–1972), as fully described in the compendium edited by Liebowitz. This approach depends on the determination of a range of fracture mechanics parameters, full details of which are decribed by Paris & Sih (1965) and Brown & Sorawley (1966).

A fracture mechanics parameter which is of particular importance in this approach is the stress intensity factor K_I and its critical value K_{IC}, which exists when K_I reaches a critical value. At this critical value a normal stress at some location in the vicinity of a crack tip causes rupture to occur; the higher the value, the less brittle the material. It can be shown that

$$K_I = \sigma Y \sqrt{a}$$

where a = crack tip dimension, Y = geometry correction factor, σ = applied stress.

The measurement techniques available for the determination of the value of K_{IC} have been reviewed by Evans (1974), who also comments on the relative advantages and disadvantages of the various specimen geometries used. The various geometries include: tapered cantilever beam, double cantilever beam, three (or four) point bending, constant moment, compact tension, double torsion.

The choice of test, and its associated specimen geometry, depends on the rate of testing, the temperature of the test, the ease of formation of the specimen (this is especially important with reference to stressing method and notch formation) and the degree of porosity of the specimen, where the flaw dimensions are critical. Consideration of method and specimen geometry is particularly important in assessing the properties of pharmaceutical materials, because the specimens tested are usually formed by compaction. Within the specimens, therefore, discontinuities or pores will exist where particles have failed to join to one another, either due to geometric or stress factors. These pores can act to create very high localized stresses at their tips. In fracture mechanics testing, the influence of the presence of these flaws is reduced by the formation of a dominant flaw in the form of a notch.

Mashadi & Newton (1987 a, b) have reported the application of four-point bending of notched beam specimens for the determinaion of the value of K_{IC} of Avicel and sorbitol. This procedure suffers from the disadvantage of having to pre-notch the specimen and has also been criticized by Evans (1974) as not being entirely satisfactory for porous specimens. For this latter type of specimen, Evans (1974) considers that the double torsion method first described by Outwater (1966) and developed by Kies & Clark (1969) is more appropriate, as it also eliminates the need to measure the crack length. Having established that pharmaceutical powders can be compacted to form specimens suitable for

Correspondence to: J. M. Newton, The School of Pharmacy, University of London, 29–39 Brunswick Square, London WC1N, 1AX, UK.

testing by this technique, this approach to measuring the value of K_{IC} for compacts is reported here.

Theory

The specimen used is a rectangular plate (Fig. 1) which is supported on four hemispheres. The load is applied by two hemispheres attached to the upper platen (Fig. 2). A groove along the length of the specimen will help guide the crack and ensure that it remains confined within the groove.

The double-torsion specimen can be considered to be two elastic torsion bars, each having a rectangular cross section such that when loaded, will exhibit small deflections. For small deflections and for specimens where the width, W, is much greater than the thickness, d, the torsion strain, θ, is given by the expression (Roark 1965)

$$\theta = \frac{Y}{W_n} = \frac{6Ta}{Wd^3g} \tag{1}$$

where T is the torsional moment $(\frac{P}{2} \times W_n)$, $\frac{P}{2}$ is the load applied to the one bar, g is the shear modulus of the material, a is the crack length and W_n is the moment arm.

On rearranging, equation 1 becomes

$$C = \frac{Y}{P} = \frac{3W_n^2 a}{Wd^3g} \tag{2}$$

where C is the elastic compliance.

Irwin & Keis (1954) showed that the strain energy release rate G is related to the compliance by the expression

$$G = \frac{P^2}{2}\left(\frac{dC}{dA}\right) \tag{3}$$

where A is the area of the crack.

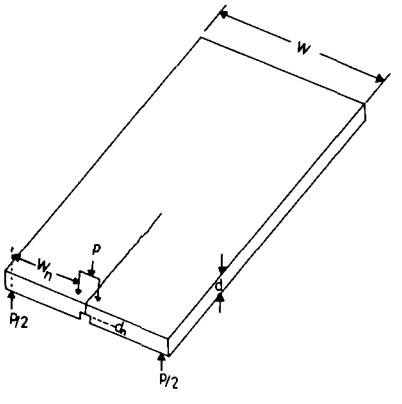

FIG. 1. Plate for double torsion method determination of critical stress intensity factor.

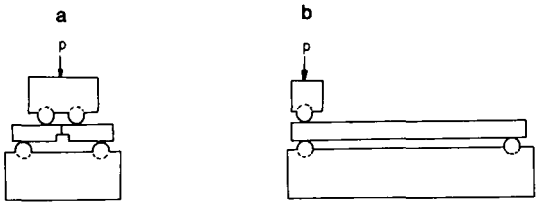

FIG. 2. Loading of plate (a) Front view. (b) Back view.

Assuming that the shape of the crack front is independent of crack length then the relationship between the crack area, the crack thickness d_n, and the crack length is $dA = d_n \times da$. Substituting for dA in equation (3)

$$G = \frac{P^2}{2d_n}\left(\frac{dC}{da}\right) \tag{4}$$

Differentiating equation 2 with respect to a gives

$$\frac{dC}{da} = \frac{3W_n^2}{Wh^3g}$$

Substituting for $\frac{dC}{da}$ in equation 4 gives

$$G = \frac{3P^2W_n^2}{2Wd^3d_ng} \tag{5}$$

The stress intensity, K, is related to G by

$$K = (EG)^{\frac{1}{2}} \tag{6}$$

Where E is the Young's modulus, so

$$K_1 = \left(\frac{3EP^2Wn^2}{2Wd^3d_ng}\right)^{\frac{1}{2}} \tag{7}$$

But E and g are related by:

$$g = \frac{E}{2(1-\gamma)}$$

Where γ is the Poisson's ratio.

Substituting for g in equation 7 gives

$$K_1 = PW_n\left(\frac{3(1+\gamma)}{Wd^3d_n}\right)^{\frac{1}{2}} \tag{8}$$

(For plane stress)

Therefore, the stress intensity factor is independent of the crack length and is a function of the applied load, specimen dimensions, and Poisson's ratio only.

Experiments with double torsion specimens have indicated that the crack front during propagation is curved and extends further along the lower face of the specimen. The analytical stress intensity for the specimen is, however, obtained by assuming a flat through crack.

Materials and Methods

Avicel PH-101 (FMC) and sorbitol 'Instant' (E. Merck) were used as received. The rectangular plates $125 \times 62 \times d$ mm (where d is the thickness of the plates) were produced by filling the die with $37{\cdot}4 \pm 0{\cdot}1$ g and $54{\cdot}2 \pm 0{\cdot}1$ g of Avicel and sorbitol, respectively. The punch and die set were placed between the platens of the Tangyes Press (Tangyes Ltd, Birmingham). The depth of the groove in the specimen was 2 mm. Four identical plates were produced at varying maximum upper punch pressures (monitored by a calibrated load cell) for each material. The large size of the specimen limited the pressure of compaction. Thus the porosity of the compacts was somewhat greater than that which can be produced by conventional tableting. The plates were stored in an air-tight sealed container for 14 days before testing.

The porosity of the plates was calculated by measuring the dimensions ($\pm 0{\cdot}01$ mm) and weight ($\pm 0{\cdot}0001$ g) of each plate and relating the volume calculated from dimensions to

that calculated from weight and density (determined by air comparison pycnometry (Beckman Model 930)). The double torsion technique was used to determine the critical stress intensity factor. Controlled pre-cracks were generated in the specimen by pre-loading the specimen at a rate of 0·025 mm min^{-1} with an Instron testing machine (Instron Ltd, model TT-CM) until a "pop in" was observed. A "pop in" or decrease in the load was used as an indication of crack growth. After pre-cracking was achieved the specimen was loaded at a rate of 0·05 mm min^{-1} until almost instantaneous crack propagation severed the specimen. This was taken as a measure of the value of P.

Results and Discussion

The experimental technique proved to be capable of providing a reproducible value for the load to cause failure. For a given formation pressure of the specimen, the coefficient of variation of the breaking load was less than 5%. From the value of breaking load, the dimensions of the specimen, and assuming a Poisson's ratio of 0·3 for all specimens, the value of K_{IC} was calculated from equation 8. The values of K_{IC} for both materials were found to increase linearly with increasing compaction pressure (Fig. 3) and with decreasing specimen porosity (Fig. 4). This indicates that as the powder becomes more consolidated, it becomes less brittle, being able to absorb greater loads before failure. The feature which allows this to occur is presumably the reduction in pore size which takes place as the powders are compacted. The process of compaction, as previously mentioned, is complex. Volume reduction, (which results in the decrease in porosity) with compaction pressure; is considered to occur by particle rearrangement, elastic deformation, plastic deformation and particle fragmentation. All these mechanisms will result not only in a reduction in the porosity of the compact but a reduction in the dimensions of the pores between the particle. From studies involving isostatic compaction of iron, Bockstiegel (1961) suggested that pores were eliminated in strict order of size, namely, the largest pores being removed first. The removal of the largest pores will remove cracks which are the most likely to allow propagation to occur when the specimen is stressed. Davidge & Evans (1970) predicted from fracture mechanics, that the largest defect (pore, inclusion,

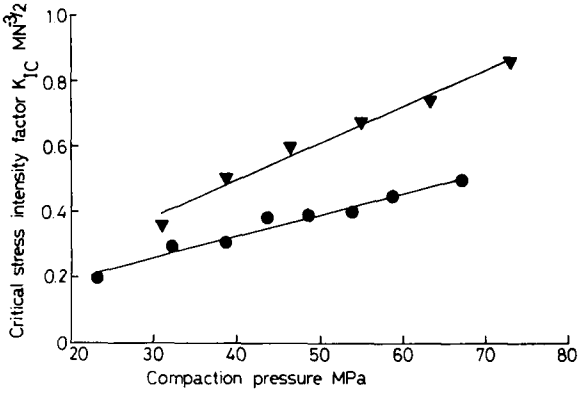

FIG. 3. Critical stress intensity factor of ▼ Avicel PH101 and ● sorbitol 'instant' as a function of compaction pressure.

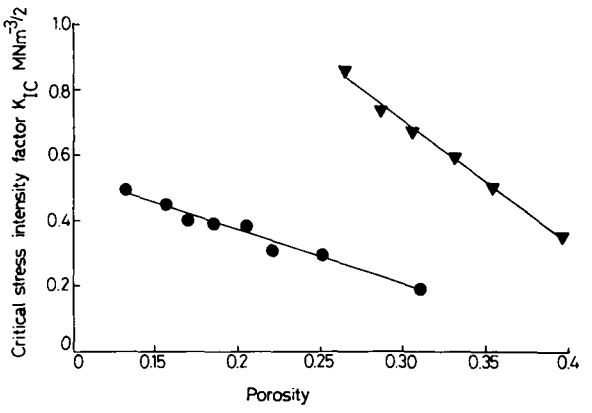

FIG. 4. Critical stress intensity factor of ▼ Avicel PH101 and ● sorbitol as a function of compaction pressure.

etc.) controls the strength of a specimen, not the overall defect concentration. Evans (1974) however, warns of the problems associated with the interpretation of results from specimens in which the defect size, in the case of compacts, the pores, are smaller than the grain size. He suggests that in this case, defects smaller than the grain size will propagate initially at the single crystal value of K_I and grain size defects are likely to propagate at K_I values intermediate between single crystal and polycrystalline values.

By assuming that all the interparticulate pores (or defects) are removed at zero porosity, the value of K_{IC} for the material can be predicted and hence the relative brittleness of the material. For the two materials tested, the linear relationship between K_{IC} and porosity allows extrapolation to zero porosity by least squares linear regression analysis. The values of K_{IC} for Avicel, PH 101 and sorbitol 'Instant' obtained by such a procedure are 1·81 and 0·69 MN $m^{-3/2}$ with standard deviations of 0·037 and 0·069, respectively.

This indicates that Avicel is basically less brittle than sorbitol 'Instant'. At equivalent values of porosity this is always the case, although the same porosity must not be taken as an indication that the pore size of the specimens is the same. The results in Fig. 4 also clearly show that the rate of change in the value of K_{IC} with decreasing porosity is greater for Avicel than sorbitol, indicating that the latter shows less sensitivity to changes in porosity than the former.

Compared with the value of 1·21 MN $m^{-3/2}$ and 0·47 MN $m^{-3/2}$ K_{IC} at zero porosity for Avicel and sorbitol beam specimens measured by Mashadi & Newton (1987a, b) the values reported here are higher. This could be caused by several factors. The double torsion method requires a value for Poisson's ratio, γ, which is not required by the beam bending system. In the current work the value of γ chosen was 0·3, a value suggested in the case for organic materials by Hiestand & Smith (1984). Church (1984) found that the value of γ for Avicel compacts can vary between 0·03 and 0·42 depending on the thickness of the beam. Using the range of γ, the value of K_{IC} for Avicel could be as low as 1·61 MN $m^{-3/2}$ or as high as 1·89 MN $m^{-3/2}$. It is also suggested (Evans 1974) that the values of K_{IC} determined from the double torsion technique are generally greater than those from notched specimens because the analysis for the former is thought to

contain a factor K_{II}, a stress intensity factor due to a shearing mode. This, therefore, is additional to the stress intensity factor due to the opening mode which is the only mode present in most notched specimen techniques. In addition, the porosity of the double beam specimen is estimated from its dimensions in which it is assumed that the porosity is uniform throughout, including the groove in which the crack propagates. This may not be the case. While the double torsion system involves this assumption, the notched beam bending is not free from assumptions, e.g. the need for an empirical geometric correction factor, and the difficulties of producing a reproducible notch. Another factor which has to be considered in the case of double beam specimens is that there is generally a greater extent of extrapolation to obtain zero porosity due to the higher porosity values of the specimen. This arises from the large specimen dimensions which require high forces to provide compaction. Porosities of less than 0·05 can be obtained for microcrystalline cellulose with normal tablet dimensions but forces of the order of 700 KN representing the limits of the press only provided porosities of the order of 0·25. That there is high correlation coefficient and a low residual variance indicates that the extrapolation will give a reasonable estimate of the value of K_{IC} at zero porosity even if the value is not absolute. It is, however, clearly essential to state the method of determination when quoting values for K_{IC} and it is necessary to compare the values for different materials determined by the same procedure if an exact comparison is required.

The values for both methods place Avicel and sorbitol 'Instant' on a scale which indicates a brittle rather than a ductile material. To establish where other pharmaceutical materials can be placed on this scale will require further work. Of equal value would be an assessment of how such factors as particle size and formation conditions influence the values of K_{IC}.

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