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BRITTLE–DUCTILE TRANSITIONS IN DIE COMPACTION OF SODIUM CHLORIDE

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Abstract—Both plastic and brittle deformation have been observed in sodium chloride particles during die compaction, with a transition from brittle to ductile behaviour occurring at a particle size of 33 μm. A reasonable agreement between data obtained from compaction measurements and those predicted from independent measurements for both yield stress and fracture toughness was found for two strain rates (0.01 and 17 s^{-1}).

INTRODUCTION

Sodium chloride is a material that is often die compacted in the pharmaceutical industry to manufacture tablets for the maintenance of the osmotic tension of blood and other cellular tissues.

It has long been considered that sodium chloride particles primarily undergo plastic deformation during compaction (Hardman and Lilley, 1970; DeBoer *et al.*, 1978) and only to a minor extent show fragmentation (Duberg and Nystrom, 1982). However, Down (1983), using scanning electron microscopy, showed that 250–500-μm crystals, with some as small as 150 μm, underwent localised fracture during compaction.

These apparent contradictions are not surprising since Badrick and Puttick (1986) have shown that for sodium chloride there is a brittle–ductile transition in the particle size range of 20 microns. Below this size, sodium chloride particles will deform plastically, whereas particles much larger will tend to crack.

In this paper we have studied the die compaction of sodium chloride over an extended range of particle sizes to show that this transition between plastic and brittle behaviour can be detected in powder compaction tests and that it is also influenced by the rate of compaction.

THEORY OF BRITTLE–DUCTILE TRANSITIONS

After an initial phase of consolidation involving particle rearrangement and settling, powder compaction generally proceeds by one or more of three distinct modes of deformation, i.e. elastic, plastic and brittle. In the case of elastic deformation the average deformation stress, σ, is governed by the elastic modulus, E, of the particles. However, for plastic flow σ depends on the yield stress σ_y and for brittle fracture it is dictated by the toughness, K_{IC}, and the particle size, d.

Generally, the plastic and brittle deformations are dominant during powder compaction of materials such as sodium chloride, so that the equations for the deformation stress are:

$$\text{plastic} \quad \sigma = \sigma_y \tag{1}$$

$$\text{brittle} \quad \sigma = AK_{IC}/\sqrt{d} \tag{2}$$

where A is a constant depending on the geometry and loading system. For compression of rectangular samples with long cracks $A = \sqrt{32/3}$ (Kendall, 1978), but for other geometries A varies (Puttick, 1980).

It is evident from these equations that large particles will crack because the stress required for brittle fracture is less than that needed for plastic flow. Small particles, on the other hand, will be plastic because the plastic deformation stress is lower than the brittle deformation stress. The transition from ductile to brittle behaviour occurs at a critical size given by

$$\sigma_{\text{plastic}} = \sigma_{\text{brittle}} \tag{3}$$

i.e.

$$d_{\text{critical}} = \left(\frac{AK_{IC}}{\sigma_y}\right)^2. \tag{4}$$

In powder compaction a specific method used to evaluate the average stress of a material during compression relies on the observations of Heckel (1961), who found that for materials that plastically deform the relative density of a material, D, could be related to the compression pressure, P, by eq. (5):

$$\ln\left(\frac{1}{1-D}\right) = KP + B \tag{5}$$

where B and K are constants.

This equation has been reappraised by Hersey and Rees (1970) who suggested that the reciprocal of K can be regarded as numerically equal to the mean yield stress of the powder. Furthermore Roberts and Rowe (1987) have shown that, for certain ductile metals and polymers, there is a good agreement between this calculated stress and the yield stress σ_y of the material as measured by indentation. Thus this method can be used to determine the plastic flow stress σ_y that is occurring during purely plastic deformation. However, in the case of a material where both plastic deformation and/or brittle fracture is occurring, it is necessary to modify eq. (5) to

$$\ln\left(\frac{1}{1-D}\right)=\frac{P}{\sigma_d}+B \tag{6}$$

where σ_d is now defined as the deformation stress of the particles, be it a plastic deformation stress, a fracture deformation stress or a combination of the two.

EXPERIMENTAL

Materials

Sodium chloride [vacuum dried, containing sodium hexacyanoferrate(II), an anti-caking agent] was obtained from Chemicals and Polymers Ltd, ICI, Runcorn. To obtain a range of sizes down to 32 μm the material was sieved using a sieve shaker. Additionally small fractions between 105 and 20 μm were obtained by grinding the 355-μm fraction in a mortar. Various fractions of the ground material were obtained by sieving using the sieve shaker and an air jet sieve. Median particle sizes were calculated from the sieve fraction data.

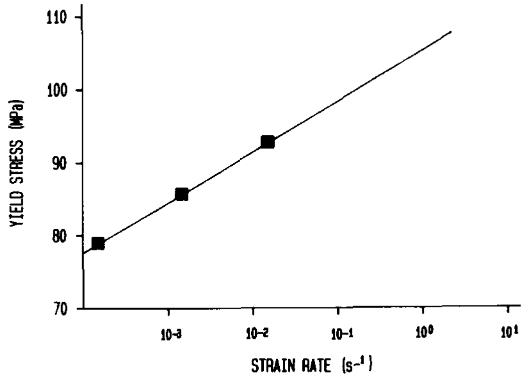

Fig. 1. Effect of strain rate on the yield stress as determined from Vickers hardness measurements of sodium chloride.

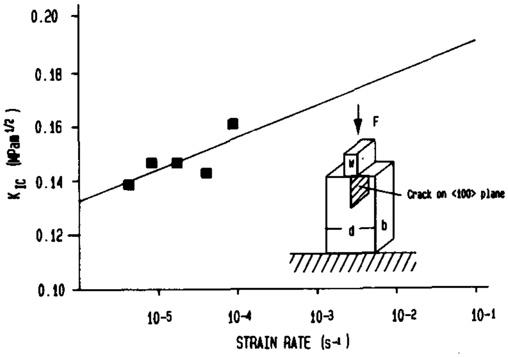

Fig. 2. Effect of strain rate on the critical stress intensity factor, K_{IC}, of sodium chloride.

Yield stress measurement

To test the above equations, it is necessary to independently measure the yield stress σ_y and the toughness K_{IC} of the salt crystals. Because sodium chloride is anisotropic in its plasticity, with a yield stress of only 2 MPa in the ⟨100⟩ direction but much higher in the ⟨110⟩ and ⟨111⟩ directions, experimental determination of σ_y was carried out by testing the Vickers hardness (Zeiss microhardness tester) of melt grown single crystals of salt (BDH) in the ⟨110⟩ direction and taking σ_y=hardness/3 (Tabor, 1951). The effect of the strain rate on the yield stress was also determined by indenting the crystal at various rates (Fig. 1).

Fracture toughness determination

The fracture toughness of sodium chloride was determined by pre-cracking a single crystal plate (BDH) in the ⟨100⟩ direction using a scalpel blade pressed into the surface, then propagating the crack by loading a 1 mm wide steel ram onto the surface using an Instron testing machine (Fig. 2). The toughness K_{IC} was calculated from the equation (Kendall, 1978)

$$K_{IC}=\frac{(1-w/d)F}{b(2d/3)^{1/2}}. \tag{7}$$

By loading the plate at different speeds, the variation of K_{IC} with strain rate was determined (Fig. 2).

Compaction

Compaction of the various size fractions was carried out using the ICI High Speed Compression Simulator (Hunter et al., 1976) fitted with 10-mm circular flat faced punches. Uniaxial compression was carried out using a single "saw-tooth" displacement–time profile for the top punch, whilst the lower punch remained stationary. Punch velocities of 0.033 and 100 mm s^{-1} were examined to evaluate the effect of the strain rate. The amount of material required to give a 3.5 mm thick compact at zero porosity was calculated from the true density (2.15 g cm^{-3}, by air comparison pycnometry). The die was cleaned after each compression with methanol and lubricated with a suspension of 2% (w/w) magnesium stearate in carbon tetrachloride to eliminate frictional interference in the measurement of the deformation stress. Measurements of displacement and force were determined to an accuracy of ±10 μm and ±0.025 kN, respectively. The elastic deformation of the punch and ram assembly were evaluated independently to allow true powder displacements to be determined. The mid straight line portion range of the plot of ln (1/1 − D) vs pressure was determined mathematically using suitable software which determined the single and double derivatives, and thus gave the pressure range of the constant slope from which the deformation stress could be determined using eq. (6).

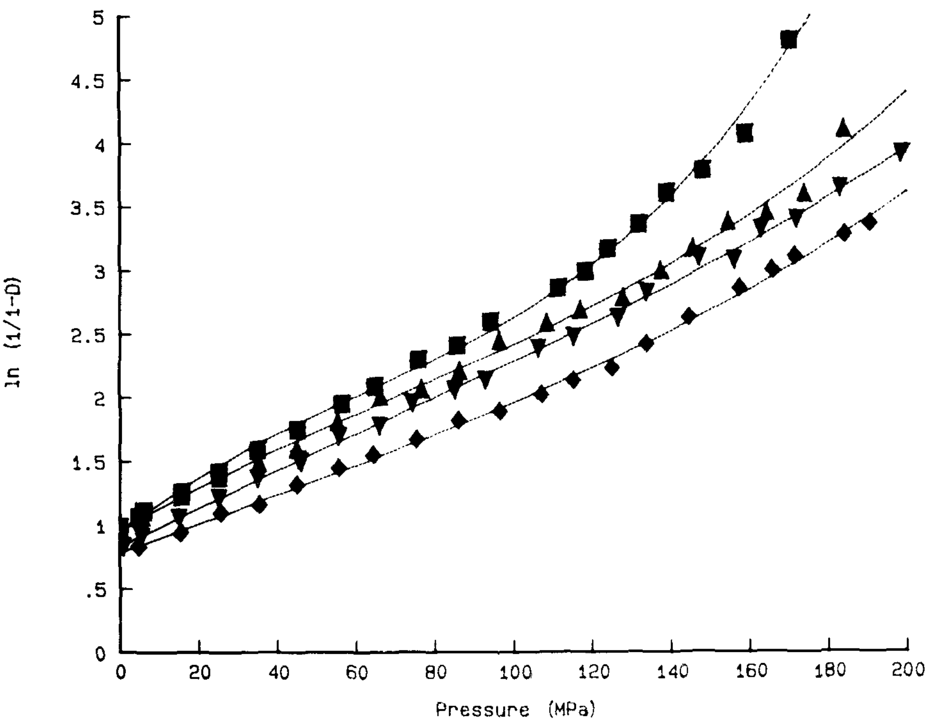

Fig. 3. Typical Heckel plot of sodium chloride for particle sizes of 328 μm (■), 116 μm (▲), 64 μm (▼) and 26 μm (◆).

Surface evaluation

Four compacts were prepared from sodium chloride crystals with median sizes of 328, 116, 64 and 26 μm using the Simulator at the slowest punch velocity. After sputter coating with gold the surfaces were examined by scanning electron microscopy to evaluate whether fracture or flow had occurred during compaction.

RESULTS AND DISCUSSION

The results for the measurement of the yield stress as determined using the Vickers indenter are shown in Fig. 1. Extrapolation of the graph (Fig. 1) to strain rates of 0.01 and 17 s^{-1}, which correspond to punch velocities of 0.033 and 100 mm s^{-1} on the Simulator, gave values of $\sigma_y = 90$ and 112 MPa, respectively. In a similar way values of K_{IC} for the same strain rates were determined by using Fig. 2, and were found to be 0.18 and 0.22 MPa $m^{1/2}$ for strain rates of 0.01 s^{-1} (0.033 mm s^{-1}) and 17 s^{-1} (100 mm s^{-1}), respectively. Chaudhri *et al.* (1981) reported a value of 0.30 MPa $m^{1/2}$ for very high strain rates for the impaction of glass and tungsten spherical particles on single crystals of sodium chloride and suggested that this was an intrinsic value, since it involved little plastic flow during cleavage. This compares favourably with that determined here for a strain rate of 17 s^{-1} and indicates that particle toughness for this compaction rate has not reached the plateau level.

Figure 3 shows typical Heckel plots for sodium chloride of various particle sizes compacted at 0.033 mm s^{-1}. The reciprocal of the slopes over the middle pressure range give the deformation stresses for the various particle sizes and are plotted in Fig. 4 as a function of grain size.

It can be clearly seen that not only for the slow punch velocity but also for the fast punch velocity the relationship between the deformation stress and grain size is extremely complex being constant for the smaller sizes below approximately 35 μm but then decreasing almost exponentially for the larger sizes. Evidence that this behaviour is consistent with a material undergoing plastic deformation at the low particle sizes and then fragmenting at the larger

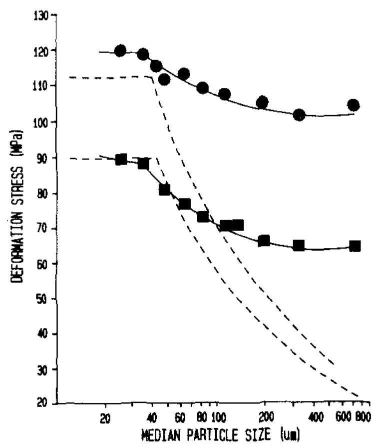

Fig. 4. Effect of particle size on the deformation stress at punch velocities of 0.033 mm s^{-1} (■) and 100 mm s^{-1} (●). The dashed lines represent the theoretical curves.

particle sizes can be obtained from two sources. Firstly, from direct observation using scanning electron microscopy (Fig. 5) which shows that tensile cracking does indeed occur for the large particle sizes while for particles 26 μm in diameter there is no evidence of cracking, and indeed evidence of plastic deformation (no evidence of cracking was seen in the crystals prior to compaction). Secondly, the values of the deformation stress for 26 μm particle size crystals compacted at the two punch velocities are comparable with the yield stresses determined from indentation, i.e. 89 and 120 MPa compared to 90 and 112 MPa, respectively. Having accepted that the inflexion in the deformation stress/particle size curve is indicative of a brittle–ductile transition, it is possible to compare the experimental results with predictions using eqs (1) and (2). Figure 4 shows the comparison using $\sqrt{32}/3$ as the constant A in eq. (2) for the two strain rates. Qualitatively agreement between the theory and the experiment is reasonable, especially at the low particle sizes,

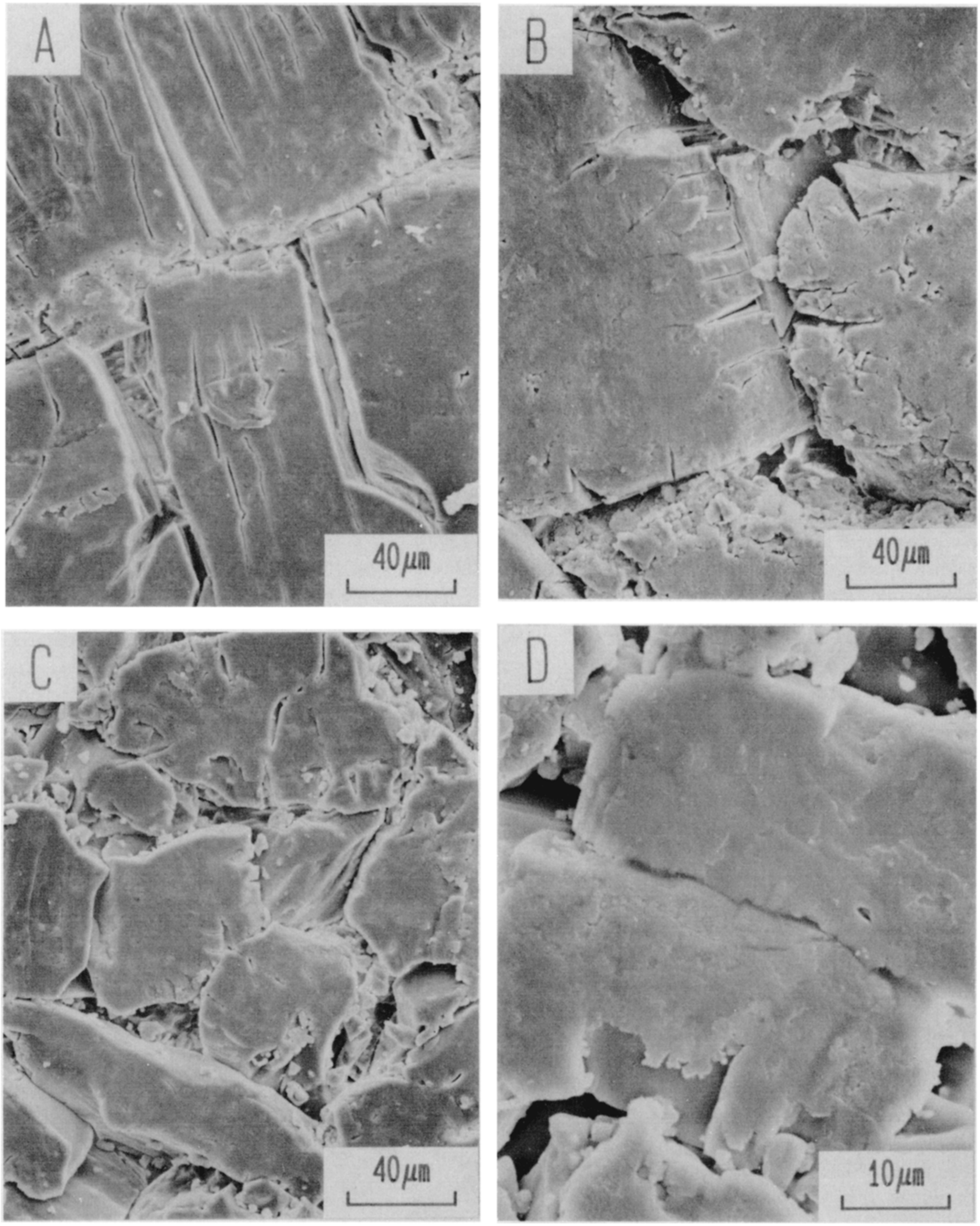

Fig. 5. Scanning electron photomicrographs of tablet surfaces for particle sizes of 328 μm (A), 116 μm (B), 64 μm (C) and 26 μm (D).

with the prediction of the critical size of 43 μm as opposed to 33 μm for the low punch velocity and 35 μm as compared to 40 μm for the higher punch velocity. At the larger grain sizes there was a more pronounced decrease in stress for the theoretical curve compared to that from the experiment. This difference is not surprising in view of the simplifications of the brittle theory which assumes a simple rectangular particle containing a single long crack in simple compression. In reality, as evident from the scanning electron photomicrographs (Fig. 5), there are many odd shaped particles in a complex state of stress with each particle containing a multiplicity of short cracks. It is also known that for sodium chloride cracking is concomitant with large amounts of plastic flow (Puttick and Badrick, 1987). Therefore the brittle equation as used here requires some modification to account for these differences between the experiment and the theory.

CONCLUSIONS

Both plastic and brittle deformation have been observed in sodium chloride particles during die compaction. The reasonable agreement between the data obtained by compaction measurements and those predicted from independent measurements for both yield stress and fracture toughness imply that it should be possible to calculate fracture toughness values for materials directly from compaction measurements over a wide particle size range.

NOTATION

A	test geometry factor
b	breadth, m
B	constant in Heckel equation
d	particle size, m
$d_{critical}$	critical particle size, m
D	relative density
E	elastic modulus, GPa
F	cracking force, N
K	constant in Heckel equation, MPa^{-1}
K_{IC}	critical stress intensity factor, MPa $m^{1/2}$
P	compaction pressure, MPa
w	platen width, m
σ	average deformation stress, MPa
σ_d	deformation stress from the Heckel plot (i.e. the reciprocal of K), MPa
σ_y	yield stress, MPa

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