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Acoustic emission of pharmaceutical materials during compression

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Summary

The acoustic emission of lactose, sodium chloride, microcrystalline cellulose and paracetamol was monitored during compression, using an acoustic transducer coupled to a portable activity meter. Three stages were identified in the compression cycle. During the application of low forces (particle rearrangement) all the materials emitted extensively. With higher forces, during particle consolidation, the samples tended to be acoustically quiet. A significant exception was that of paracetamol, which did not exhibit a quiet stage through the whole compression cycle. This high level of energy dissipation indicated that little of the available compression energy was actually used for particle bonding, resulting in the poor compression properties well known for paracetamol. Acoustic emissions were again high during the third, post-compression stage. The extent of emission was dependent on tablet lubrication and the material being compressed. For sodium chloride and a free flowing lactose, a decrease in particle size produced a corresponding reduction in emmisive counts, indicating that as particle size is reduced the number of fractures reduces proportionally. The results suggest that acoustic emission measurements may enable the brittleness of tabletting materials to be quantified and aid the assessment of compression characteristics.

Introduction

The technique of acoustic emission measurement is popular in both the disciplines of ceramics and metallurgy for the non-destructive testing of materials. The use of the technique in pharmaceutical technology has, however, been limited to work by Rue et al. (1979) for the detection of capping during tabletting and by Rue and Barkworth (1980) for the monitoring of stress relaxation in sodium chloride tablets. Work was also undertaken by Board (1980) to try to establish a link between acoustic emission and tablet fracture during the hardness test. Acoustic emissions are used by metallurgists to establish the deformation mechanisms of metals under stress and it has been suggested by Brown (1979) that acoustic emissions from brittle materials are different from that of ductile materials.

This study was undertaken to monitor the acoustic profiles of common pharmaceutical materials during compression. The materials selected bonded by different mechanisms and the work was initiated in order to establish if these mechanisms could be classified using acoustic emission.

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Materials and Methods

The materials examined in the study were as follows: Zeparox E.P. Lactose (Dairy Crest Whey Division), sodium chloride Analar grade (BDH Chemicals), Lactose BP (Dairy Crest Whey Division) Avicel PH101 microcrystalline cellulose (F.M.C. Corp.), paracetamol Analar grade (BDH Chemicals). Both the Zeparox and sodium chloride were sieved into 5 particle size fractions: 200–250 μm, 160–200 μm, 125–160 μm, 63–125 μm and 32–63 μm. 100 g of each powder was sieved for 20 min at amplitude 4 using a Fritsch vibrating sieving shaker (Model 3.502). An Instron Universal testing machine (Type 1121, 10 kN load cell) was fitted with a specially designed tablet compression assembly which ensured alignment of the upper punch when entering the die. Conventional F machine tablet punches were fitted to the assembly. A horizontal hole was drilled through the die-holder to permit the fitting of an acoustic wave guide. Fig. 1 shows how the wave guide was spring loaded against the side of the die. Acoustic coupling gel was used to enhance the acoustic signal from the die to the wave guide. PVC sleeving was placed around the wave guide inside the die support table to reduce attenuation of the acoustic signal. The acoustic signals were detected using a type D140B acoustic transducer (Dunegan U.K.) which was fitted to the end of the wave guide. This is a differential type of transducer which gives a high common mode rejection rate in order to reduce interference from electromagnetic sources. The transduced acoustic signals were conditioned using a Portable Activity Monitor (P.A.M.) Model 4103 (Dunegan U.K.). Fig. 2 shows a block diagram of the instrumentation. The bandwidth of the P.A.M. was internally selected from 95 kHz to 600 kHz. The P.A.M. has internal switches for selection of a series of acoustic parameters. The parameters monitored in this study included:

Total Counts = the total number of acoustic emissions which crossed the preset threshold; *Total Events* = an acoustic event is recorded when no emissions were detected crossing the threshold during 1 ms; and *Count Rate / Event Rate* = the rate at which counts or events occur were also measured.

The optimum sample period for count and event rate in this trial was found to be 1.0 s due to the limitations of the recording system.

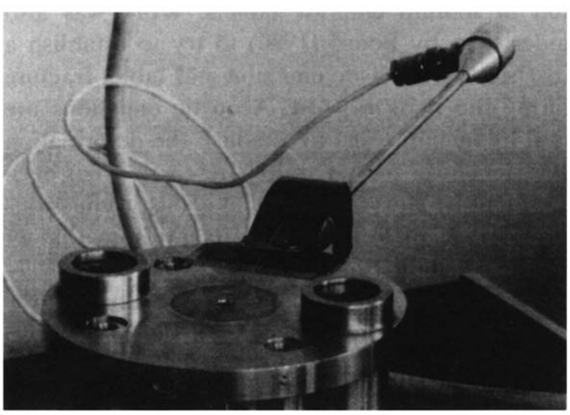

Fig. 1. Wave guide spring loaded against the side of the die.

Average signal level

A secondary output enabled the continuous monitoring of the average signal level (A.S.L.) to be measured in dB. Outputs from the P.A.M. were recorded on a two pen Euroscribe Chart Recorder (Model B5217).

Monitor

The acoustic emission signal was monitored using a transient recorder (Datalab DL 902) coupled to an oscilloscope (Telequipment DM 63). The sample rate on the transient recorder was set to 1 μs giving a total sample time of 2 ms. The stored transient signal was then recorded on the pen recorder at an output rate of 100 ms per sample.

All the compressions were made to an applied load of 270 MPa at a compression speed of 10 $mm \cdot min^{-1}$ using 1/4 inch flat-faced punches. Tablet weight for all materials was 225 mg. The die was lubricated between each compression with a 10% solution of stearic acid in chloroform; the solution being applied with a cotton bud.

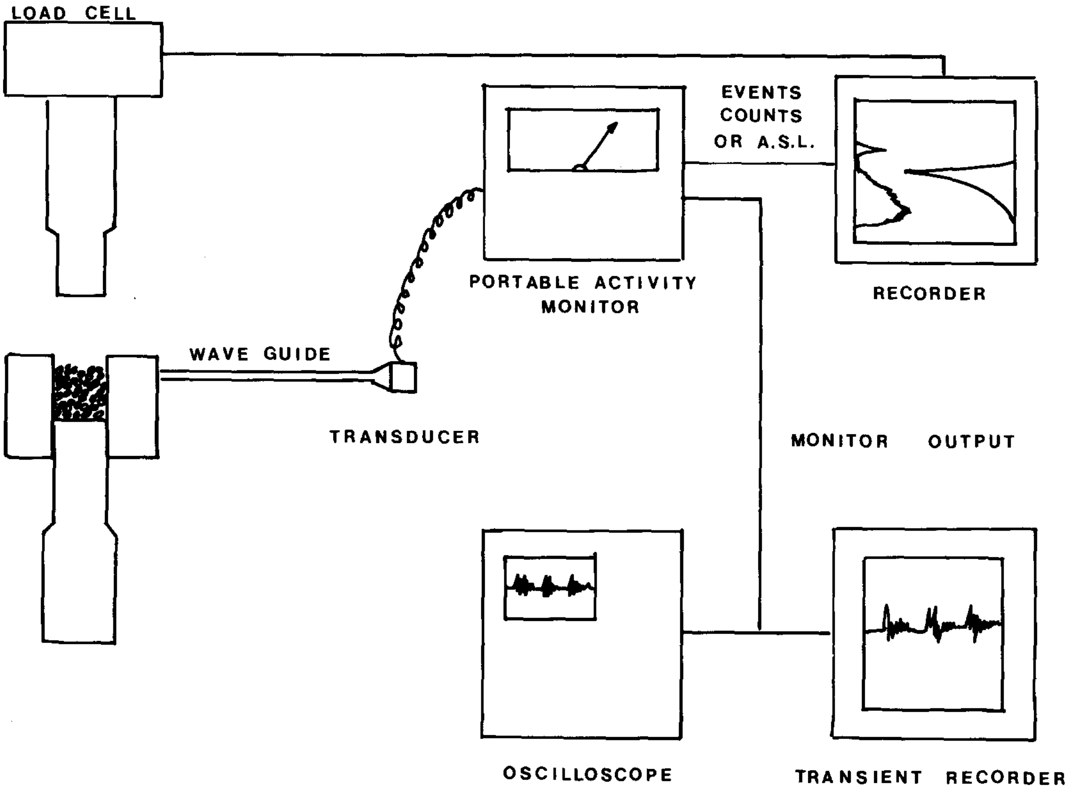

Fig. 2. Instrumentation for acoustic emission.

Results and Discussion

Average signal level

A.S.L. was monitored for all the materials at all stages of the compression cycle. Although the modulus of the amplitude differed for each material, a pattern of the signal levels during the compression cycle became evident. The three stages identified during the compression cycle are shown in Fig. 3 and are described as follows.

(1) Reorganisation or rearrangement. During the application of low forces, all of the materials tested had the highest amplitudes during this stage. The maximum amplitude of the rearrangement peaks for the sodium chloride was 5–6 times greater than for the Zeparox. As can be seen from Fig. 4 a reduction in particle size of both the sodium chloride and Zeparox lowers the reorganisation peak of the A.S.L.

(2) The compression or consolidation stage. The majority of the samples tested tended to be acoustically quiet during this stage although peaks were often observed in the smallest particle size range of the Zeparox. Subsequently, peaks at this stage of the cycle were shown to occur during the compression of lactose B.P. and Zeparox in an unlubricated die. However, peaks did not occur at this stage with unlubricated sodium chloride.

(3) The third stage was a post-compression peak which was present with all the materials tested. This peak increased in magnitude with consecutive unlubricated compressions in the case of lactose B.P., indicating a post-compression lubrication effect, as the tablet relaxed after compression. The ratio of this peak to the rearrangement peak was much higher in the case of Zeparox compared with sodium chloride.

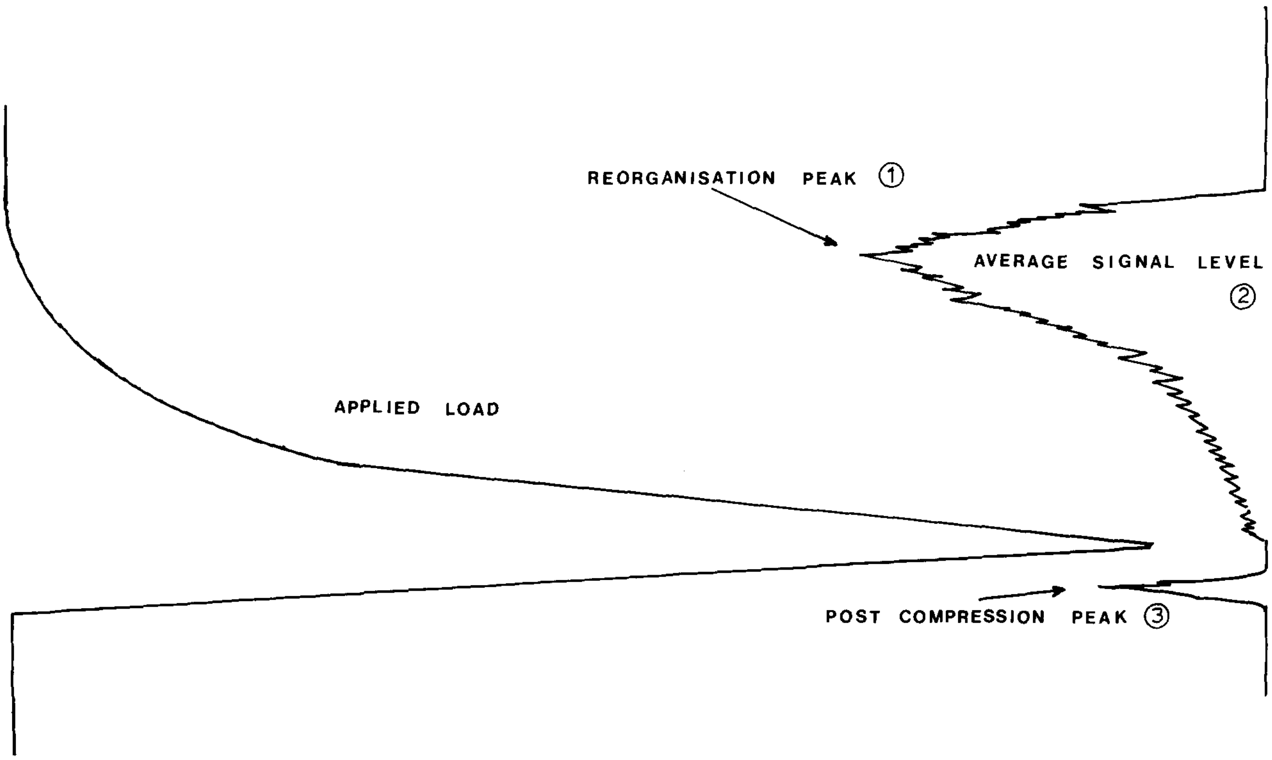

Fig. 3. Acoustic profile for sodium chloride 250–200 μm.

Total counts / events

When the meter was set in a totalising mode the counters would hold the current value measured until either more counts were detected or the meter was reset. This was also the case for the count output to the chart recorder making it possible to measure total counts in mV using the linear scale on the recorder and converting this to counts

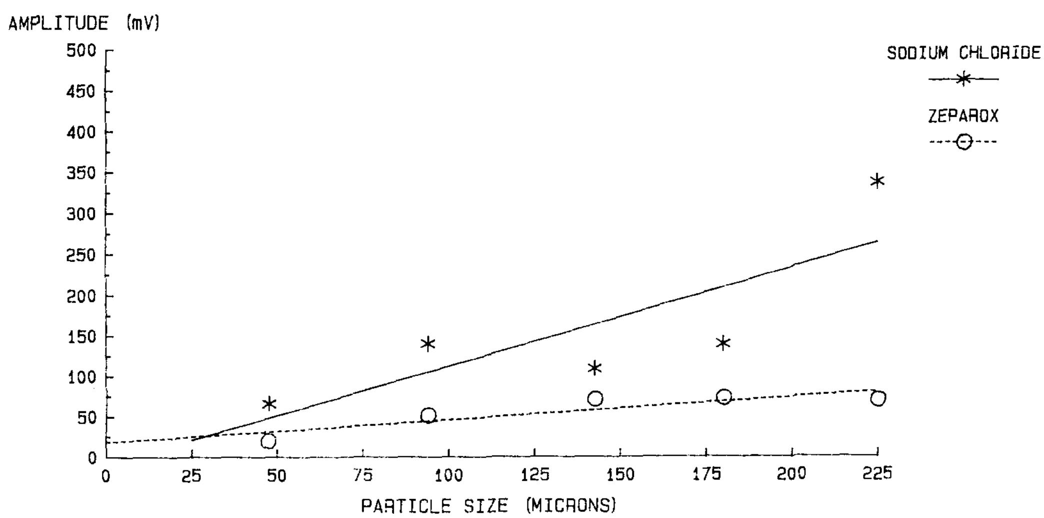

Fig. 4. Average signal level: reorganisation peak.

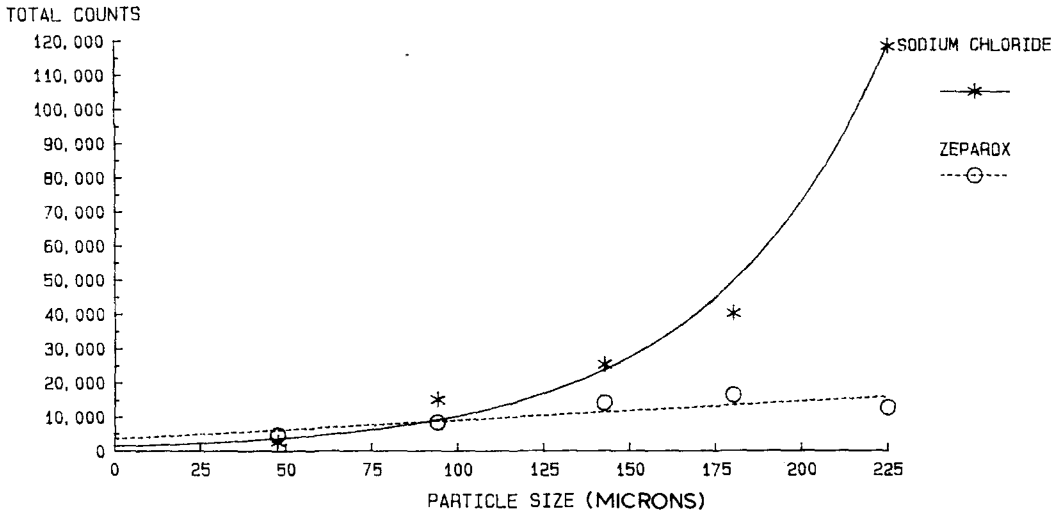

Fig. 5. Effect of particle size on total counts.

or events using a converting equation. The total number of counts was calculated by this method because of the poor resolution of the meter on the P.A.M. Nevertheless, as the chart scale of 0–1000 mV represented a log scale of $0–10^6$ counts, large variations in counts were observed for relatively small variations in mV reading. Fig. 5 shows, however, that it was still possible to resolve differences in total counts for individual materials and for a range of particle sizes of the same material. Total counts exhibited a similar trend to that of A.S.L., that is, a reduction in the total number of counts with smaller particle sizes indicating that the number of fractures is related to the number of emissions. The total counts for different materials were ranked in the same order as for the maximum A.S.L. value observed during the rearrangement stage. Paracetamol having the greatest number of counts followed by sodium chloride, Zeparox and lactose B.P.

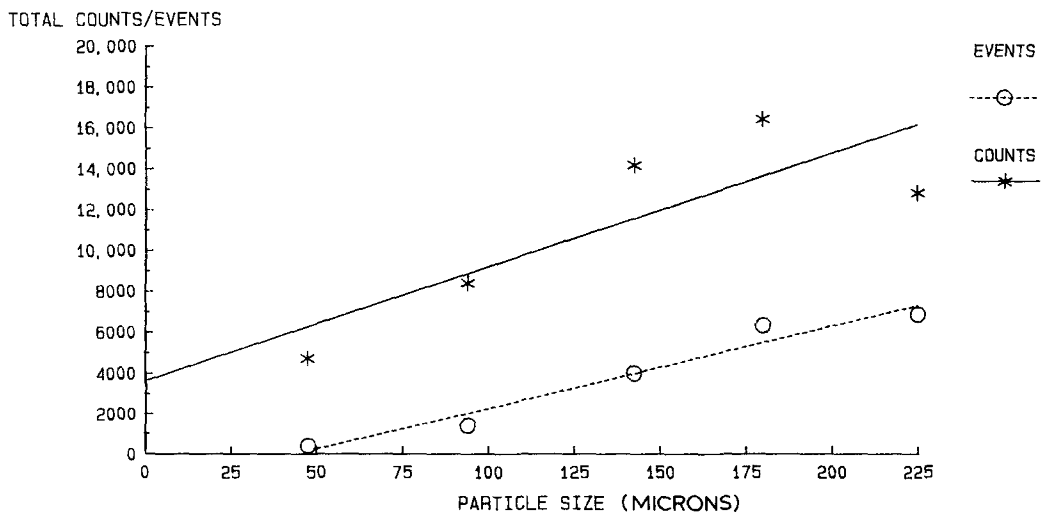

Fig. 6. Zeparox – effect of particle size on counts and events.

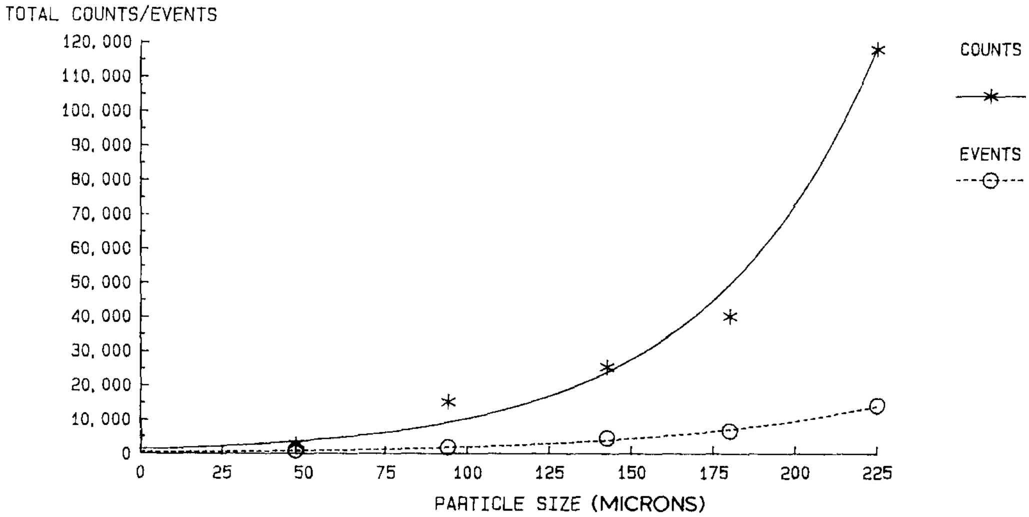

Fig. 7. Sodium chloride – effect of particle size on counts and events.

Acoustic events showed a similar trend to counts in that a reduction in the number of events occurred with smaller particle sizes of the same material. As can be seen from Figs. 6 and 7 no obvious pattern exists in the ratio of counts to events for the different particle size ranges although generally the number of counts/events were lower for Zeparox than for sodium chloride, which may simply be due to a feature of the higher number of counts for sodium chloride or as a result of the more ordered collapsing of the sodium chloride crystals under stress.

Count rate / event rate

This measurement mode was appropriate for observing the count levels during the progression

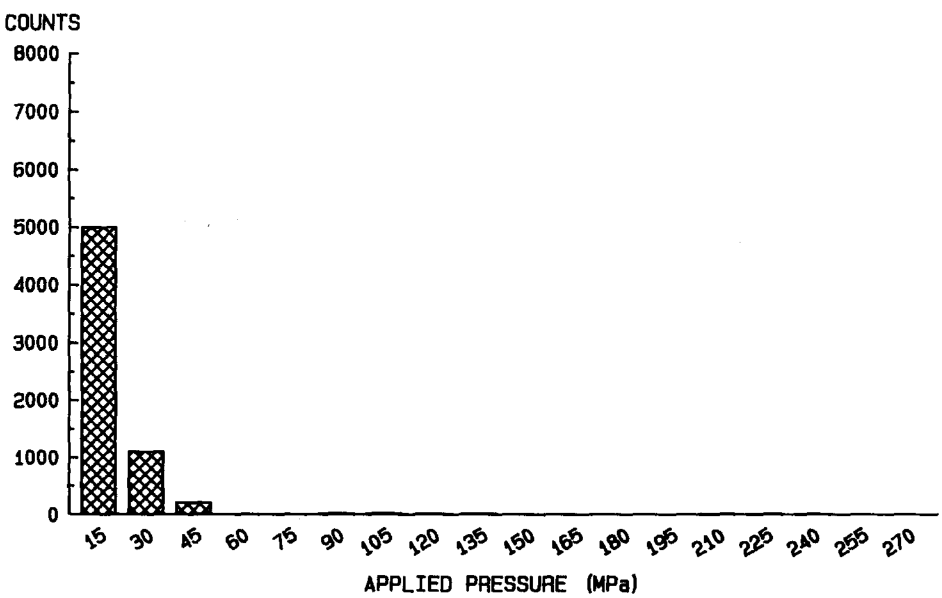

Fig. 8. Applied pressure vs count rate for sodium chloride.

of the compression cycle. The P.A.M. was set at a 1 s sample rate to keep within the dynamic fidelity of the recorder, producing a histogram type of plot of counts/s. When plotted against compression force similar patterns to the A.S.L. were observed for Zeparox and sodium chloride, both materials showing a quiet stage after rearrangement. Sodium chloride showed reduced emissions at about 60 MPa and Zeparox at about 15–18 MPa. The count rate plotted against load for sodium chloride is shown in Fig. 8. The emissions from sodium chloride may be the result of brittle fracture occurring at low forces whilst above 60 MPa plastic deformation undetected by the transducer appears to be the dominant factor. Evidence for brittle fracture in sodium chloride at low loads was observed by Down (1983).

As can be seen from Fig. 9, paracetamol did not exhibit a quiet stage during the whole compression cycle and although the count rate reduced towards the maximum compression pressure of 270 MPa, emissions were still occurring at this level. As the smaller particle size fraction of both Zeparox and sodium chloride were considerably quieter than the larger fractions, paracetamol with a mean particle size of 45 μm would have expected to be acoustically quiet; however, it was shown to be extremely noisy during compression. The high number of counts may be interpreted as a high level of energy being dissipated indicating that little of the compression energy being applied is used for tablet bonding, resulting in the poor compression properties well known for paracetamol. It also suggests that a highly brittle mechanism is present.

It is very difficult to quantify the contribution made by the die-wall/powder interface to the acoustic emissions and to establish that the emissions observed are particle/particle interactions. However, as can be seen from Fig. 10, as all materials exhibited a higher number of counts during compression compared with ejection, it appears likely that additional phenomena other than the particle/die wall interface are contributing to the emissions recorded. Compression cycles both without powder and with Avicel were shown to be acoustically quiet. This demonstrates that as yet it may not be possible to detect plastic deformation emissions in pharmaceutical materials using this technique with this type of instrumentation, primarily because of the low amplitudes likely to be involved in plastic deformation and the attenuation of these signals across the die. The technique may, however, be useful for classi-

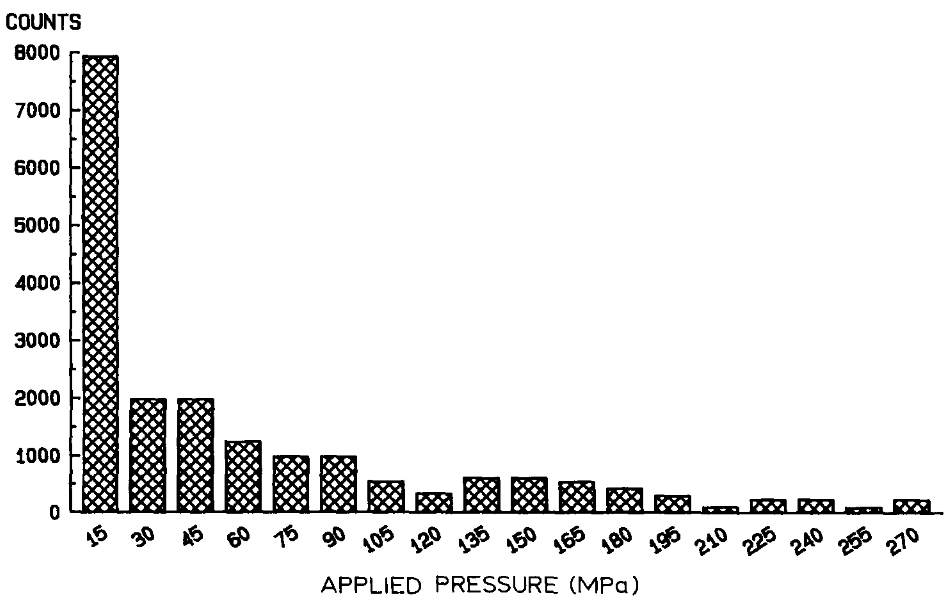

Fig. 9. Applied pressure vs count rate for paracetamol.

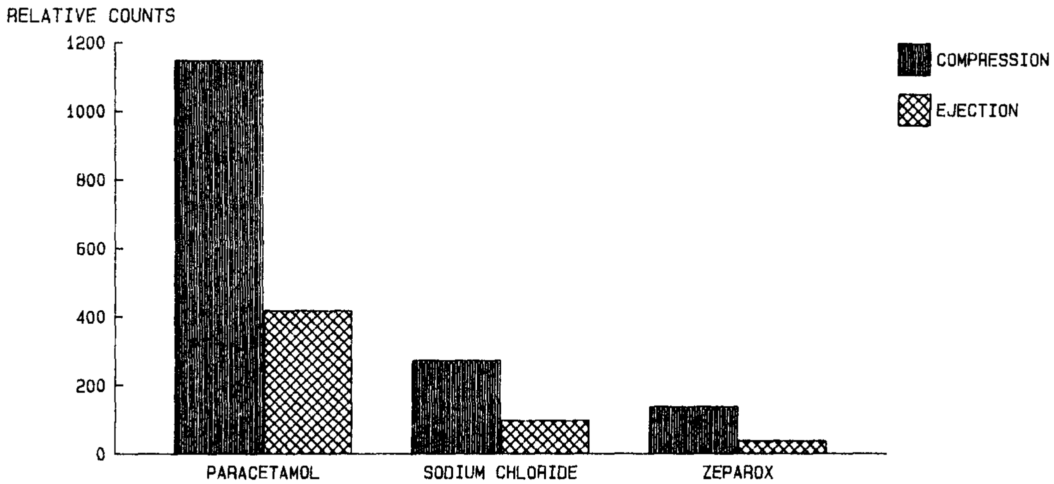

Fig. 10. Comparison of compression counts with ejection counts.

fying the brittleness of materials under compression and in combination with other techniques for lubrication evaluation. Although sophisticated techniques may be required, an analysis of the frequency content of the emissions may also be a useful method of classifying materials during compression.

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