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Acoustic emission during the deformation of α-lactose monohydrate and anhydrous α-lactose monocrystals

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Abstract—During the deformation of single crystals of α-lactose monohydrate and anhydrous α-lactose in a crushing strength rig, their acoustic activity was monitored using a portable activity meter. The acoustic parameters measured were the average signal level (ASL), count rates and total acoustic counts. Both types of lactose, even though deformed by fragmentation, differed fundamentally in the degree and nature of this fragmentation. Close correlation was observed between the ASL, count rate profiles and the force-displacement profiles. The monohydrate form is acoustically more active than the anhydrous form during deformation. Small internal fractures which were neither visually observed nor detected in the force-displacement profiles (in particular the anhydrous α-lactose) were detected by monitoring the acoustic signals during the deformation of these crystals. This work illustrates the potential of using the acoustic emission technique as an aid in the assessment of the deformation characteristics of pharmaceutical materials during single crystal compression studies.

Studies on the bulk compression of lactose have shown that anhydrous α-lactose behaves differently from α-lactose monohydrate. The anhydrous form has been shown to exhibit greater compressibility when compared with the monohydrate form (Lerk et al 1983; Vromans et al 1985). In our previous study, assessments of the fundamental mechanical and compression characteristics of these two lactose types were carried out using single crystals (Wong et al 1988). These crystals were assessed by microindentation and compressive deformation. These techniques provide both visual and quantitative confirmation of their bulk compression characteristics. However, there is increasing need to have a better insight into the deformation and fragmentation process of crystals during compression.

There is interest currently in the use of the acoustic emission technique for the evaluation of pharmaceutical materials (Waring et al 1987a, b, c). As part of a continuing effort in the study of the mechanical and deformation behaviour of single crystals of pharmaceutical interest, this work explores the prospect of using the acoustic emission technique as an investigative tool in the assessment of the fragmentation processes of α-lactose during compressive deformation of single crystals.

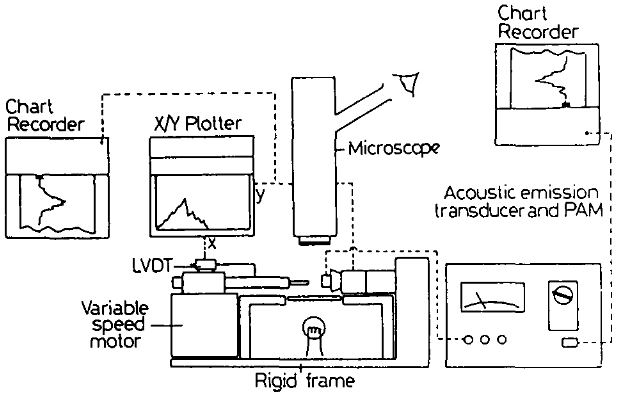

FIG. 1. The single crystal compression rig and the portable activity monitor.

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Materials and methods

α-Lactose monohydrate (Sigma, Poole, UK) was used as the starting material. Large macroscopically well-formed α-lactose monohydrate single crystals with a range of sizes were grown by an agar-gel suspension technique (Wong & Aulton 1987). Anhydrous α-lactose crystals were obtained by methanol desiccation as described previously (Wong et al 1988). Well-formed single crystals of α-lactose monohydrate and anhydrous α-lactose of size range 2·5–4·5 mm in length were individually mounted on the crystal holders using cyanoacrylate adhesive. The adhesive was allowed to cure for 7 days before mechanical testing.

Single crystals of α-lactose monohydrate and anhydrous α-lactose were subjected to compressive deformation using a compression rig developed by Wong et al (1988). The compression rig was modified by replacing the static platen with a special holder into which a ceramic-head differential type acoustic transducer (D9203A, Physical Acoustics Ltd, Cambridge, UK) was fitted. This arrangement allowed direct crushing of the crystal onto the acoustic transducer with simultaneous monitoring of the acoustic activity and the force-displacement profile (Fig. 1). The crystals were subjected to deformation at a compression rate of 8·5 μm s^{-1}.

The acoustic signals were conditioned using the Physical Acoustics/Endevco Model 4103 portable activity monitor. The operational frequency range was set internally to between 95 and 600 kHz. The count rate and total counts were monitored. The sampling period for the count rate was set at 1·0 s. A secondary output which monitored the average signal level (ASL) in dB was also recorded on a chart recorder.

Results

Representatives of the ASL profiles, count rates and force-displacement (F-D) curves are illustrated in Figs 2–5. The

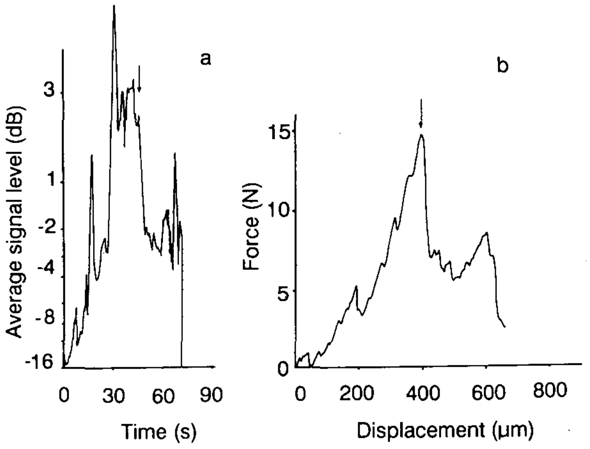

FIG. 2. (a) ASL profile and (b) force-displacement curve during compressive deformation of an α-lactose monohydrate monocrystal.

arrows on the ASL, F-D and count rate profiles indicate the point of final major fracture and hence the collapse of the crystals. Brittle materials exhibit a wide variation in fracture characteristics as a result of the random distribution of defects within their crystal structure. Therefore brittle fracture is a random event and, whilst the overall shape of the F-D and acoustic profiles are similar for each type of lactose, variation in the detail of individual curves was observed.

The shapes of the F-D curves obtained from the two types of α-lactose were different. The monohydrate crystals gave very jagged F-D curves (see Figs 2b, 4b) whereas the F-D curves of the anhydrous crystals were much smoother during compressive deformation (see Figs 3b, 5b). The state of the crystals at various points on the F-D was followed visually. Permanent records were also made by taking photographs at regular time intervals during the deformation process. Details on the qualitative analysis and quantitative assessment of the deformation process for these crystals were described in an earlier publication (Wong et al 1988).

Differences in the acoustic activity as shown by the ASL profiles were also observed for the two types of α-lactose (see Figs 2a, 3a). Only semi-qualitative analysis was carried out on the ASL profiles. For comparative evaluation, the number of peaks, both distinctive large peaks and small peaks, were noted. In general, more peaks were observed on the ASL profiles of both types of lactose when compared with those on the F-D profiles.

Analysis and comparison of the ASL and F-D profiles revealed that the large distinctive peaks on the ASL profiles were matched by the major "force peaks" on the F-D curves. This was particularly clear in the case of the monohydrate crystals. Small peaks on the ASL profiles also corresponded to small peaks on the F-D curves. In some cases, no noticeable peaks were registered on the F-D curves although it was confirmed visually that the crystals had undergone spalling fractures. Fig. 2, shows that the ASL profile for the α-lactose monohydrate crystal followed the F-D profile, showing many steep rises and falls.

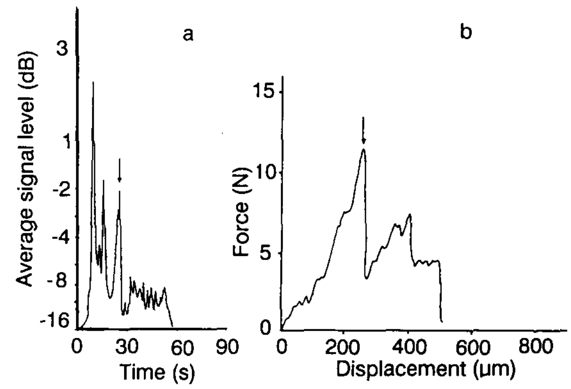

FIG. 3. (a) ASL profile and (b) force-displacement curve during compressive deformation of an anhydrous α-lactose monocrystal.

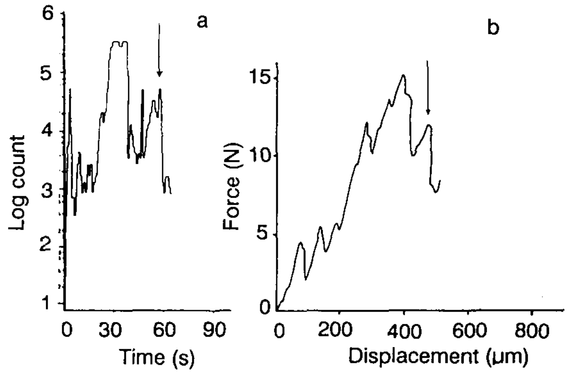

FIG. 4. (a) Count rate profile and (b) force-displacement curve during compressive deformation of an α-lactose monohydrate monocrystal.

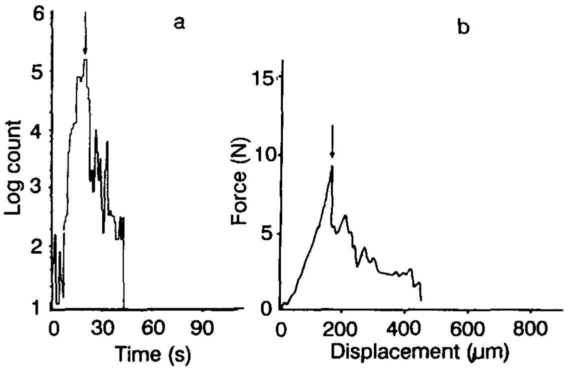

FIG. 5. (a) Count rate profile and (b) force-displacement curve during compressive deformation of an anhydrous α-lactose monocrystal.

For anhydrous crystals, in addition to the large ASL peak associated with the final fracture (as seen in Fig. 3b), abrupt changes in ASL were also frequently detected although no apparent peak was registered on the F-D curves. Fig. 3a shows a jagged ASL profile compared with the relatively smooth F-D curve (Fig. 3b).

The count rate profiles of α-lactose monohydrate and anhydrous α-lactose are shown in Figs 4a and 5a, respectively. Again, the count rate profiles for both types of lactose are similar to that of the F-D profiles. From the count rate profiles, the total acoustic counts up to the point of main fracture and collapse of the crystal was measured. Fig. 6 shows the total acoustic counts up to the point of main fracture for a range of crystal sizes of both types of lactose. The result clearly shows that the monohydrate form is acoustically more active than the anhydrous form during compressive deformation.

Discussion

In our earlier publication (Wong et al 1988), we reported that anhydrous α-lactose is more deformable and mechanically weaker than the monohydrate crystals. If these data were to be considered in isolation, the increased deformability of the anhydrous form may be misinterpreted as the occurrence of ductile deformation, which was clearly not the case. Both types of α-lactose were observed to undergo deformation by fragmentation but differed in their fragmentary behaviour. This difference was also reflected in the shape of the force-displacement profiles (see Figs 2b–5b). The very jagged force-displacement curve of the monohydrate crystal is directly due to the intermittent fragmentation caused by propagation of spalling cracks through the crystal and separation of fragmented pieces from the main crystal. On the other hand, the anhydrous crystal deformed by gradual localized crushing at the point of contact without any intermittent fractures or large internal fractures. The major fracture of the anhydrous crystal corresponds to ultimate breakage of the crystal into two halves.

To further assess the deformation behaviour of these crystals, this work monitored the acoustic activity generated during the

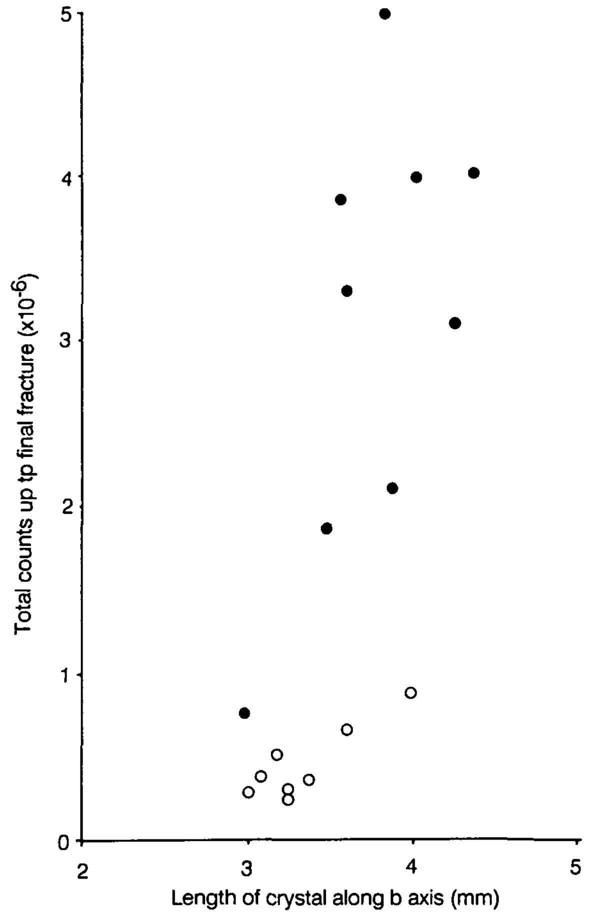

FIG. 6. Comparison of the total acoustic emission counts from the compressive deformation of monocrystals of α-lactose monohydrate (●) and anhydrous α-lactose (○).

deformation of these crystals. The abrupt changes in ASL detected during the compression of both types of α-lactose crystals is characteristic of fragmentation. In addition, fundamental differences in acoustic activity were also observed for the two types of lactose. The monohydrate form was observed to be acoustically more active than the anhydrous form. Generally, the anhydrous form emits acoustic signals of lower magnitude (as reflected in the ASL and count rate profiles). Furthermore both the number of ASL peaks and total acoustic counts recorded for the anhydrous crystal were also much lower. These observations confirmed our previous study which suggested that these two types of α-lactose differ in both the nature and the degree of fragmentation during compressive deformation. The level of acoustic activity also suggested a lower energy involvement for fragmentation during the deformation of the anhydrous lactose, thus further complementing earlier work which showed that the anhydrous form is mechanically more deformable and weaker than the monohydrate crystal.

This work also showed that fractures which were otherwise not detected either by monitoring the F-D curves or visual observation could be picked up by monitoring the acoustic activity emitted during the deformation process. This is clearly illustrated by the increase in the number of noticeable peaks on the ASL profiles when compared with that of the F-D profiles (see Figs 2, 3). This is particularly true for the anhydrous crystal as illustrated by Fig. 3. Since acoustic emissions represent elastic energy generated from the relief of stresses when a body undergoes deformation and fracture, the high ASL detected in the anhydrous crystals well before the main fracture (indicated by the arrow) suggested the occurrence of internal fractures which were not confirmed visually and only detected as small peaks on the F-D profiles. The lack of visual evidence is attributed to the opaque nature of the anhydrous crystal. The low force registered for these internal fractures can be attributed to the fact that these fractures were small and of low elastic energy (as confirmed by the low ASL level and acoustic counts) and the high deformability of the anhydrous crystal may have absorbed the compressive force below the sensitivity of the load transducer. Therefore the correlation of acoustic emission with a force displacement profile provides a clearer insight into the fragmentation process which takes place within a crystal during compression.

This work shows that α-lactose monohydrate crystals which fracture by spalling cracks emit more acoustic signals than anhydrous α-lactose crystals which undergo fragmentation by crumbling without any propagative fracture. In addition, the acoustic emission technique can provide valuable information about what is taking place within the crystals during the deformation of single crystals, thus complementing data generated from force-displacement profiles and visual evidence obtained during the deformation of single crystals. Studies of this nature are important in providing a better understanding of material behaviour during bulk compression.

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