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The surface hardness of tablets

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A pneumatic micro-indentation apparatus has been used to determine the hardness of tablets of aspirin at various points on their diameter. All the tablets were 13 mm diameter; the compaction pressure and the particle size of the initial material were varied. The compression was carried out in a Perspex die so that radial pressure could also be assessed during compaction. The micro-indentation apparatus measured the total deformation at the loaded point on the tablet, and also the elastic recovery when the load was removed.

The strength, porosity, abrasion resistance and ease of disintegration of a tablet vary over its surface and through its thickness. By compacting magnesium carbonate in a 2 inch diameter die with gelatin-encapsulated manganin strain gauges buried in the powder, Train (1957) showed that a distribution of pressure existed. The pressure distribution was closely paralleled by the distribution of density as determined by machining and weighing the compact. Shear at the surface of a tablet is known to produce a denser skin, and Train & Hersey (1960) demonstrated this effect in compacts made from lead shot; good adhesion was obtained only where shear had been intense. By increasing the compaction pressure, the density of tablets is increased (Higuchi, 1954; Lewis & Train, 1965). The friability decreases, and the disintegration rate and penetrability by water or solvents also decline (Ganderton, 1969).

Aspirin on compaction can be work-hardened (Ridgway, Glasby & Rosser, 1969) and it thus seemed possible that, by making aspirin tablets and assessing their hardness at various points on their surfaces, it might be possible to show that non-uniformity persists even in comparatively small compacts in which it might not be expected.

EXPERIMENTAL

The aspirin was a crystalline product (Laporte Industries Limited). Four fractions were obtained by sieving: 20–30 mesh, 30–40, 40–44 and 44–60, mean particle sizes 670, 460, 388 and 303 μm respectively.

Weighed quantities were compacted by a hydraulic press with steel punches in a 13 mm diameter Perspex die of the type described by Ridgway (1966), which when viewed by polarized light gave an interference fringe pattern from which the radial pressure at the die wall could be estimated. Fringe patterns were photographed for subsequent examination. Tablets were ejected from the die with more difficulty than from steel dies, but were of good quality and adequate for subsequent testing.

Two series of tests were made, in each of which one parameter was held constant whilst the other was varied; the two parameters being compaction pressure and particle size. The tablet weight remained constant within each series. Five indentations were made over each test area and the average value used in the plotting of the graphs. The test areas were five concentric regions of equal width on the tablet face.

Point hardnesses were determined using a pneumatic micro-indentation apparatus [Research Equipment (London) Ltd.] that was originally developed for the testing of paint coatings. It consists of a spherical sapphire indenter, diameter 1·55 mm, which can be lowered on to the test surface under a selected load of a few grams. The depth of the indentation and the recovery when the load is removed can both be measured, the timing of the loading–unloading cycle being automatically programmed. The indenter movement is measured by a double pneumatic amplification system of the flapper and nozzle type and is displayed on a pneumatic recorder, on which the full scale deflection corresponds to 6 μm indenter movement. A typical chart record is shown in Fig. 1. ABC is the loading curve, the vertical

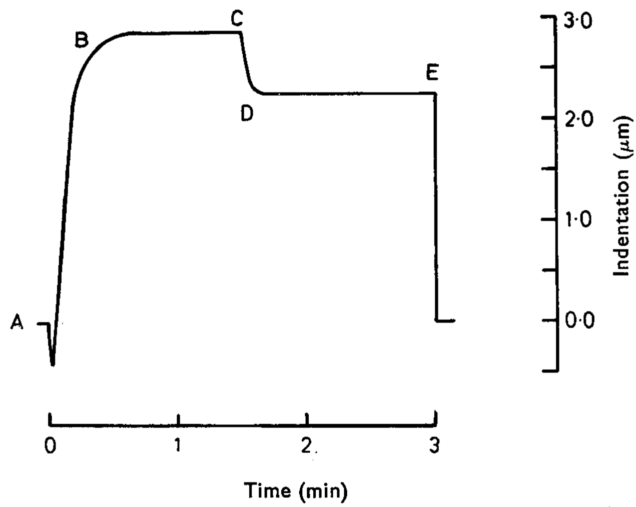

FIG. 1. The curve produced by the pneumatic recorder during the indentation of a tablet. The abscissa is time, the indentation and relaxation taking about 1½ min each. The ordinate is the depth of penetration of the indenter into the tablet, the maximum deflection representing 6 μm. ABC is the penetration under load, and CDE the recovery on removal of the load.

distance AC being the depth to which the indenter penetrates. CDE is the recovery curve followed when the load is removed at C, so that CE is the elastic recovery of the specimen from the indentation. The vertical distance AE is the plastic deformation or permanent set, being the depth of the indentation which remained in the tablet surface. This is the depth which would be measured, inferentially, in a conventional pyramidal diamond-indentation test where the diameter of the impression is measured by a microscope and graticule. The depth of the impression is usually a fifth of the length of its diagonal. As the pneumatic indentation test measures both elastic and plastic components of the deformation, the test is more useful than the normal microhardness test. A further advantage is that the indentation size is measured and displayed, so that inaccuracies due to the poor visibility of the indentation in a microscope graticule do not arise.

The thickness of each tablet was measured with a micrometer. In conjunction with the weight and the diameter, this enabled the density of the compact to be found.

At each level of compaction pressure and particle size listed in the tables of results, one tablet was made. At each radial position, five determinations of hardness were made. For both elasticity and hardness, the standard deviation was about 20%. The measurement technique is much more precise than this: almost all the experimental scatter is caused by the intrinsic variability of the material under test.

RESULTS AND DISCUSSION

We first make some comments on the pneumatic hardness test. As the spherical sapphire indenter is lowered slowly onto the tablet surface, the following stages occur.

The tablet first deforms elastically according to the equation first deduced by Hertz (1881). The area of contact, a circle of diameter d, follows the law

$$\frac{d}{2} = \left\{\frac{3}{4}\,Wr\left(\frac{1-\sigma_1^2}{E_1} + \frac{1-\sigma_2^2}{E_2}\right)\right\}^{\frac{1}{3}} \quad \ldots \quad \ldots \quad (1)$$

where W is the applied load, r is the indenter radius, E_1 and E_2 are the Young's moduli of the tablet and the indenter, and σ_1 and σ_2 are the corresponding Poisson's ratios. As more of the total load able to act on the indenter is progressively transferred to it by the micro-indentation apparatus, W is effectively increasing continuously during loading. The distribution of pressure and stress in and below the circular area of contact between the indenter and the tablet is not uniform. The pressure is a maximum at the centre, where the value $P_{max} = 3/2\ P_{mean}$, P_{mean} being defined as $W/{}^{\pi}/_{4}\ d^2$. The pressure falls from the centre to the periphery of the indentation according to the equation

$$P_x = P_{max}\ \{1 - x^2/(d/2)^2\}^{\frac{1}{2}} \quad \ldots \quad \ldots \quad \ldots \quad \ldots \quad (2)$$

where P_X is the pressure at a distance x from the centre.

Stresses in the material beneath the indentation follow a rather complex pattern, but the maximum shear stress occurs on the centre line at a depth of about half the radius of the contact zone, and has a value of 0·47 P_{mean}, for a material having a Poisson's ratio of 0·3. Since most materials in tension yield at a shear stress of about half their yield stress, Y, plastic deformation of the material beneath the indenter will begin at a loading for which $P_{mean} = 1{\cdot}1$ Y. Plastic deformation then spreads until all the material beneath the indenter is yielding. The pressure distribution gradually changes from the parabolic so that the pressure at the edge of the indentation rises ($P_{max} \approx 3{:}5$ Y, $P_{mean} \approx 2{\cdot}8$ Y). Deviation from the parabolic distribution is small enough to ensure that, when the load is removed from the indenter, the elastic recovery reduces the depth of the impression but does not change its diameter. It is this fact which makes ordinary Brinell hardness measurements possible (see Fig. 2).

Normally, in hardness testing, the diameter of the impression is determined and, from it, the hardness is calculated by means of the formula

$$\text{Brinell hardness number} = \frac{W}{\frac{\pi D}{2}\left(D - \sqrt{D^2 - d^2}\right)} \quad \ldots \quad \ldots \quad (3)$$

where D is the diameter of the spherical indenter. The diameter of the impression is unchanged by removing the load. To determine the extent of the elastic recovery it is necessary to measure the change in depth of the indentation on unloading, and this the pneumatic micro-indentation apparatus does.

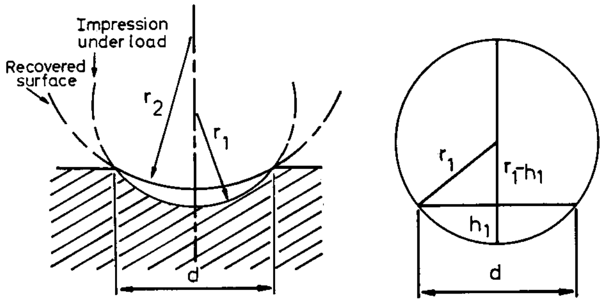

FIG. 2. The change in form of the indentation when the load is removed. Under load, the indenter, radius of curvature r_1, penetrates to a depth h_1 giving an impression of diameter d. When the load is removed, elastic recovery occurs, and the radius of curvature of the impression increases to r_2, its depth decreasing to h_2. The change, $h_1 - h_2 = \Delta h$, is a measure of the elasticity.

Referring to Fig. 2, the following equations may be obtained (Tabor, 1951):

(a) $$d = 2{\cdot}2 \left\{ \frac{W}{2} \cdot \frac{r_1 r_2}{r_2 - r_1} \left\{ \frac{1}{E_1} + \frac{1}{E_2} \right\} \right\}^{\frac{1}{3}} \quad \ldots \quad \ldots \quad (4)$$

This is the Hertz equation for the load W necessary to push the sphere, radius r_1, back into the residual spherical indentation, radius r_2, which is left when the load is removed; it is the reverse of the elastic recovery step. The Poisson's ratios σ_1 and σ_2 have been taken as 0·3.

(b) It can be seen from the geometry of Fig. 2 that $h \simeq \frac{d^2}{8r}$ provided $h \ll r$ and that

$$\Delta h = h_1 - h_2 = \frac{d^2}{8} \left\{ \frac{r_2 - r_1}{r_1 r_2} \right\}$$

where h_1 and h_2 are the depths of penetration with the load present and with the load removed. Now from the equation (4)

$$\frac{r_2 - r_1}{r_1 r_2} = \frac{2{\cdot}2^3}{d^3} \cdot \frac{W}{2} \cdot \left(\frac{1}{E_1} + \frac{1}{E_2} \right)$$

$$\therefore \quad \Delta h = \frac{2{\cdot}2^3}{8d} \cdot \frac{W}{2} \cdot \frac{1}{E_1} \quad \ldots \quad \ldots \quad \ldots \quad \ldots \quad \ldots \quad (5)$$

where E_1 is the Young's modulus of elasticity of the tablet. E_2, the Young's modulus of the sapphire indenter, is so large that $1/E_2$ is negligible. Since $h_1 = d^2/8r_1$, $d = \sqrt{8 h_1 r_1}$, and $r_1 = 0{\cdot}775$ mm, thus $\Delta h = 0{\cdot}268 \frac{W}{E_1 \sqrt{h_1}}$ or, rearranging,

$E_1 = 0{\cdot}268 \frac{W}{\Delta h \sqrt{h_1}}$, where W is in kg, Δh and h_1 in mm and E_1 is in kg/mm^2. Thus the Brinell hardness is obtainable from the initial depth of penetration using equation (3), and the modulus of elasticity from the recovery after removal of the load.

The results of the measurements are given in Tables 1 and 2. In all cases the applied pressure was sufficient to compress the aspirin practically to zero porosity.

Table 1. *Variation of tablet hardness and modulus of elasticity over a tablet face as a result of changing the compaction pressure.* Table weight = 0·5 g. Indentation load = 4 g. The tablets were compressed from aspirin of ungraded size.

Compaction pressure kg cm^{-2}	Position on tablet	Indentation depth h_1 (μm)	Elastic recovery Δh (μm)	Brinell hardness number kg mm^{-2}	Modulus of elasticity kg mm^{-2}
Surface 1: adjacent to stationary punch					
1088	1	1·46	0·54	0·56	7·7
	2	2·14	0·88	0·38	3·9
	3	2·39	0·99	0·34	3·3
	4	2·39	1·04	0·34	3·2
	5	2·62	1·21	0·31	2·6
	Av.	2·20	0·93	0·39	4·1
1453	1	1·66	0·62	0·50	6·4
	2	1·89	0·62	0·44	6·0
	3	2·19	1·14	0·38	3·0
	4	2·11	1·00	0·39	3·5
	5	1·46	0·62	0·56	6·7
	Av.	1·86	0·80	0·45	5·1
1815	1	1·46	0·69	0·56	6·1
	2	1·37	0·62	0·60	7·0
	3	1·34	0·84	0·61	5·1
	4	2·02	1·13	0·41	3·2
	5	1·81	0·85	0·45	4·1
	Av.	1·60	0·83	0·53	5·1
2183	1	1·47	0·87	0·60	4·8
	2	1·48	0·81	0·56	5·1
	3	1·52	0·82	0·54	5·0
	4	1·85	1·17	0·45	3·2
	5	1·43	0·70	0·57	6·1
	Av.	1·55	0·87	0·54	4·9
Surface 2: adjacent to moving punch					
1088	1	1·59	0·58	0·58	6·9
	2	1·39	0·54	0·59	8·9
	3	1·66	0·74	0·50	5·3
	4	2·38	1·04	0·35	3·2
	5	2·46	1·36	0·33	2·4
	Av.	1·90	0·85	0·46	5·3
1453	1	1·27	0·50	0·65	8·9
	2	1·51	0·53	0·54	7·7
	3	1·49	0·63	0·55	6·6
	4	1·82	0·95	0·45	3·9
	5	2·06	0·76	0·40	4·6
	Av.	1·63	0·67	0·52	6·4
1815	1	1·48	0·78	0·56	5·3
	2	1·33	0·77	0·62	5·7
	3	1·45	0·80	0·57	5·2
	4	1·69	0·95	0·49	4·1
	5	1·52	0·92	0·54	4·5
	Av.	1·49	0·84	0·55	5·0
2183	1	1·06	0·53	0·78	9·2
	2	1·25	0·56	0·66	8·0
	3	1·75	0·77	0·47	4·9
	4	2·17	0·85	0·38	4·0
	5	1·56	0·66	0·53	6·1
	Av.	1·56	0·67	0·56	6·5

Table 2. *Variation of tablet hardness and modulus of elasticity over a tablet face as a result of changing the particle size of the compressed material.* Tablet weight = 1·0 g. Indentation load = 4 g. All tablets were compressed at 1453 kg cm^{-2} (143 MNm^{-2}).

Particle size μm	Position on tablet	Indentation depth h_1 (μm)	Elastic recovery Δh (μm)	Brinell hardness number kg mm^{-2}	Modulus of elasticity kg mm^{-2}
Surface 2: adjacent to moving punch					
303	1	1·19	0·42	0·69	11·0
	2	1·18	0·50	0·70	9·2
	3	1·21	0·55	0·68	8·5
	4	1·07	0·49	0·77	10·0
	5	1·34	0·59	0·61	7·4
	Av.	1·20	0·51	0·69	9·2
388	1	1·35	0·59	0·61	7·4
	2	1·03	0·35	0·80	14·3
	3	1·20	0·49	0·68	9·3
	4	1·36	0·52	0·60	8·3
	5	1·49	0·51	0·55	8·1
	Av.	1·29	0·49	0·65	9·5
460	1	1·15	0·45	0·71	10·5
	2	1·47	0·59	0·56	7·1
	3	1·81	0·64	0·45	5·9
	4	1·45	0·79	0·56	5·3
	5	1·80	0·91	0·46	4·1
	Av.	1·54	0·68	0·55	6·6
670	1	1·63	0·58	0·50	6·9
	2	1·83	0·60	0·50	6·2
	3	1·86	0·55	0·44	6·8
	4	1·97	0·79	0·42	4·6
	5	2·14	0·52	0·38	6·7
	Av.	1·89	0·61	0·44	6·2

Measured tablet densities all lay between 1·284 and 1·297 g/cm^3. This 1% variation in density is not thought to be significant: compaction to zero voidage was probably achieved in all cases. Thus any variation in mechanical properties over the tablet surface cannot be ascribed to a voidage distribution since the voidage is everywhere zero. The radial pressure exerted on the die wall during each compression increased with increasing compaction pressure, but no significant difference could be found between the behaviour of the different particle size fractions.

Fig. 3(a) shows that there is an increase in the surface hardness of the tablet as the compaction pressure is increased; such behaviour is characteristic of a work-hardening substance. It has been observed previously (Ridgway, Glasby & Rosser, 1969) and was one reason for carrying out the present investigation. Provided that other factors are kept constant, increase in particle size causes the tablets to be softer; as the initial crystal size is increased from 300 to 700 μm, the Brinell hardness is linearly reduced from 0·7 to 0·4 kg/mm^2. Smaller crystals have more inter-particle contacts per unit volume of crystalline material than do larger ones, so that for a given degree of compaction there is more contact pressure and consequent work-hardening.

Fig. 3(b) shows the change of hardness across the faces of a tablet. These tablets were made by applying thrust to one punch only, the other being stationary and that

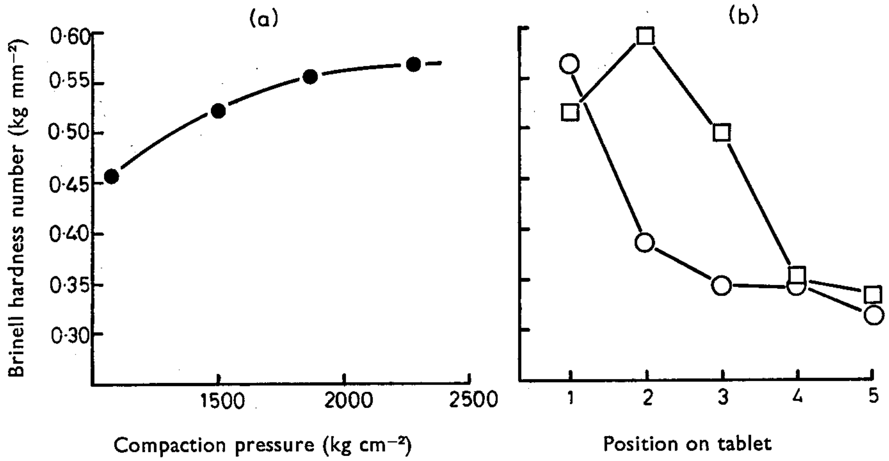

FIG. 3. (a) The average hardness of the tablet surface as the compaction pressure is increased, showing the overall work-hardening effect. (Compaction pressure range 100-250 MNm^{-2})

(b) Surface hardness of the two faces of a tablet. The centre is harder than the outer edge for both faces, and the face contacted by the moving punch is harder than that contacted by the stationary punch. —○—Stationary punch face. —□— Moving punch face. Numbers 1–5 refer to radially equally-spaced positions, 1 at the centre and 5 at the outer edge of the tablet.

side of the tablet adjacent to the moving punch is harder than the other. Also, the tablet is harder at the centre than at the edge. This conclusion is rather unexpected. In the work of Train, shearing and wedge action were thought to be greatest near the outer edge of the moving punch, as this was a region of greater pressure and resultant tablet density. Train's explanation was that the Boussinesq (1876) pressure bulb was modified by the die wall to give high pressure regions in the lower central part of the compact and in the upper edge. However, there are several differences between Train's compacts and ours. He used magnesium carbonate in much smaller particle sizes than we have used for aspirin. His compacts were large, about 2 inches across and 3 inches deep, whereas ours are much smaller overall and have a depth only about $\frac{1}{4}$ of their diameter. His applied pressures compacted his powder to about 70% of the theoretical density. Train's results were in agreement with those of Kamm, Steinberg & Wulff (1949) but not with those of Duwez & Zwell (1949).

We suggest that in small tablets the Train mechanism may be considerably modified

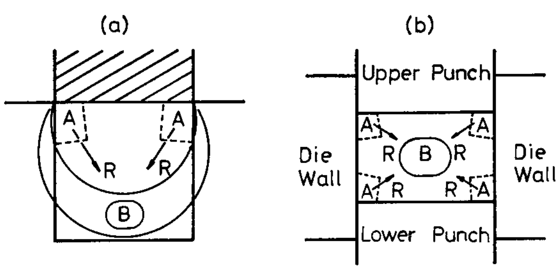

FIG. 4. (a) The Boussinesq pressure bulb, as modified by the die wall. The wedging action produces shear and a high density at A, and the forces R acting together give a high pressure and density at B.

(b) For a tablet whose depth is much less in comparison with its diameter, forces R, acting from both sides and at a greater angle to the vertical, produce a high pressure and density at the centre, with perhaps a high shear zone at the outer edge. This shear is increased if the tablet is moved relative to the die wall during compaction.

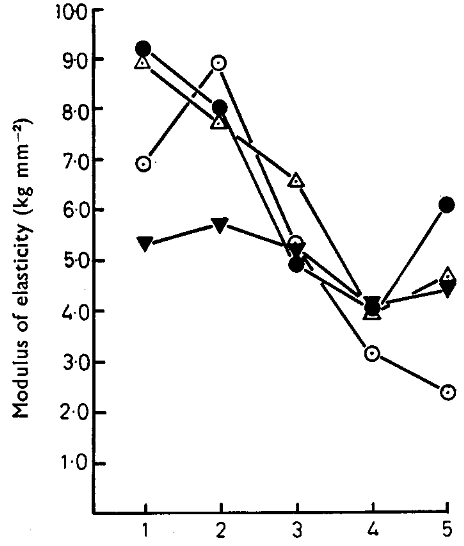

FIG. 5. Young's modulus of elasticity as a function of distance from the centre of the tablet The compaction pressures are: —⊙— 1088 kg/cm^2. —△— 1453 kg/cm^2. —▼— 1815 kg/cm^2. —●— 2183 kg/cm^2. (Range 106-216 MNm^{-2})

as shown in Fig. 4. In Fig. 4 (a), the pressure bulb modified by the die wall is shown. In Fig. 4 (b) a further stage is indicated. The resultants R of the Train mechanism are caused, by the proximity of the other punch and its reaction, to swing round at a greater angle to the vertical. The high density region found by Train near the base of his compacts will now occur at the tablet centre. In the case of aspirin there will not be a region of higher density, but a region of greater work-hardening, since

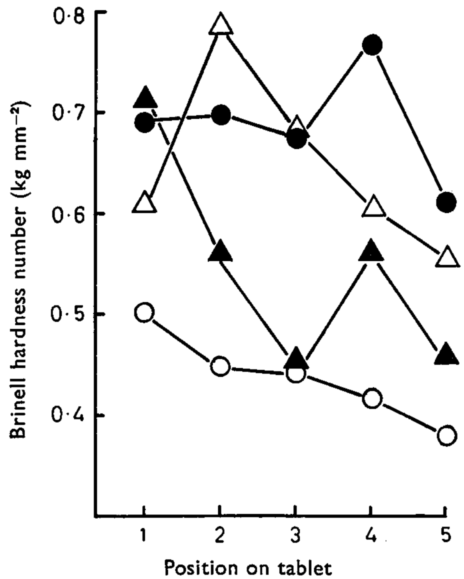

FIG. 6. The variation of hardness with position on the tablet face for different particle sizes of the initial material. —●— 44–60 mesh, mean 303 μm. —△— 40–44 mesh, mean 388 μm. —▲— 30–40 mesh, mean 460 μm. —○— 20–30 mesh, mean 670 μm.

greater applied pressure and greater hardness correlate for this substance. Using concave punch faces might well assist this process by directing pressure towards the tablet centre and enlarging the central hard region so that it formed a greater proportion of the tablet.

The elasticity for the surface adjacent to the moving punch is plotted in Fig. 5. The elasticity does not change greatly with pressure level. It is high at the centre, where the material is work-hardened and therefore able to resist further deformation and permanent set. It decreases outwardly along the radius, but shows a slight increase at the extreme outer edge. This behaviour differs from that of the Brinell hardness itself, and indicates that the effect of a hydrostatic pressure upon aspirin, such as obtains at the centre of the compact when it is made, may well be different from the effect of a shear force which occurs at points near to the edge of the punch faces. Sheared aspirin is softer, but will elastically recover better from a deformation. Compressed aspirin is physically harder, but when it deforms it does so plastically with less recovery.

Fig. 6 shows the variation of hardness with position on the tablet face for the different particle sizes used. Although the experimental scatter is large, the increase of hardness with decrease of crystal size is apparent and there is a decrease of hardness from the centre of the tablet to the outer edge.

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