

# Temperature changes occurring during the compression and recompression of solids

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Measurements of the temperature rise occurring on compression, recompression and relaxation of compacts formed from Asagran, sodium chloride and boric acid have been made by means of thermocouples inserted into the compressed materials. There are marked differences in temperature rise for the different compacts when compressed to a final force of 50 kN (5 tons). With all compacts there was a rise in temperature on initial compression and a fall in temperature on relaxation. The rise could be separated into two components—that due to compact formation plus a further rise due to its elastic compression. The fall in temperature is thought to result on release of the strain energy stored in the compact. The rise in temperature on recompression followed by the fall of relaxation could be repeated many times on the same compact. Both rise and fall were directly proportional to the final compression force used. The fall on relaxation appeared to be numerically less than the rise on recompression for compacts of boric acid and Asagran. Lubrication of Asagran with magnesium stearate had little effect on the temperature rises.

Compressed tablets examined immediately after manufacture are often warm especially if produced on a high speed rotary machine. This thermal effect seems to have attracted little attention.

Nelson, Busse & Higuchi (1955) predicted a temperature rise of about 5° C during the formation of a sulphathiazole tablet. Hanus & King (1968), using thermochromic indicators, suggested that the rise in temperature on hard compression of sodium chloride could be as much as 30° C; however, thermometric measurements on ejected tablets showed the rise to be far less.

No attempts have been made to measure temperature rise by means of thermocouples placed within the tablet, although Juslin (1969) has made use of them to measure temperature rise on the upper and lateral surfaces of tablets during compression. Some preliminary results obtained by using thermocouples within compacts are now described.

## EXPERIMENTAL

The equipment used is shown diagrammatically in Fig. 1. A commercial flat-faced punch and die assembly (1.59 cm d) was used with the die annealed so that a hole (1 mm d) could be drilled through the wall. It was then rehardened and tempered. Thermocouples were constructed of two 30 cm lengths of enamelled copper wire (0.101 mm d) bridged by one 8 cm length of insulated Eureka wire (0.127 mm d). The junctions were formed using a capacitance discharge welder (Spemibly Technical Products Ltd.).

Exactly half of a weighed quantity of the substance to be compressed was then poured into the die with the lower punch in position. One junction was pushed through the hole so that the junction was centrally placed and the remainder of the

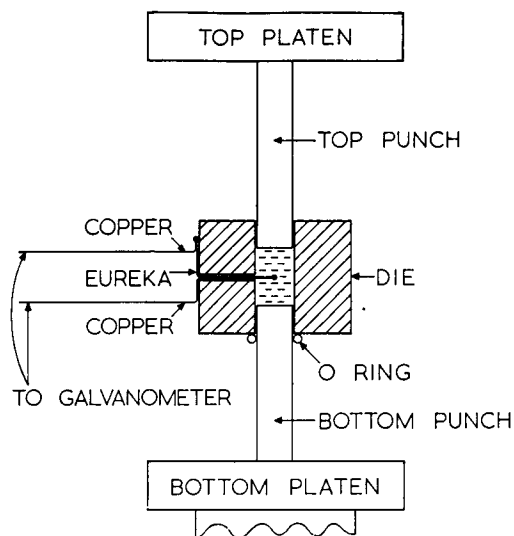


FIG. 1. The punch and die assembly.

material poured into the die. After placing the top punch into position, the assembly was placed between the platens of a C.30 manually operated hydraulic press (Research and Industrial Instruments Co. Ltd.). The remaining junction was fixed in contact with the die wall using a small piece of plasticine. The two copper wires were connected to a BB 40 galvanometer forming part of a direct recording oscillograph (Honeywell 1706 Visicorder). This symmetrical arrangement avoided the use of soldered connections or terminal blocks which can often give rise to troublesome thermal e.m.f.s. There was no detectable net e.m.f. when the copper wires were connected to a sensitive valve voltmeter of full-scale deflection  $100 \mu\text{V}$ .

Thermocouples were calibrated before use by placing the junctions in separate beakers of water at room temperature. The galvanometer spot was then made coincident with a "marker" spot provided from a mirror within the recorder. A calibration curve was constructed by raising the temperature of one beaker of water by  $1^\circ\text{C}$  intervals and plotting the galvanometer deflection against the temperature difference of the two junctions up to a final difference of  $20^\circ\text{C}$ . The calibration curve was nearly linear and had a slope of  $2.4 \text{ mm}/^\circ\text{C}$ . No differences were apparent when three such assemblies were calibrated and when thermocouples were constructed in the same way from identical lengths of wire, it was assumed that they had the same characteristics.

#### *Measurement of temperature rise on compression*

This was made when the compact was formed by compression with the recorder chart speed of  $6 \text{ mm s}^{-1}$ ; the heating and cooling curves obtained are like those in Fig. 2, temperature axes being added using the calibration graph. The undulating form of the temperature rise is due to the several strokes of the ram required to reach the ultimate compression force—usually  $50 \text{ kN}$  (5 tons). When the compact was removed on completion of the measurements, the thermocouple had usually retained its central position and it did not appear to interfere with the formation of a good sound compact.

*Thermal diffusivity of the compacts*

A preliminary estimate of the relative thermal diffusivities of compacts formed from the materials used was obtained in the following way. Compacts of the same thickness were prepared by compression to 50 kN. These were taken in turn and one junction of the thermocouple assembly glued to one face while the other junction was immersed in water at room temperature. The opposite face of the compact was pressed in contact with a hotplate at approximately  $70^{\circ} \pm 5^{\circ} \text{C}$  using a thin glass rod to maintain light pressure. A trace of the rise in temperature of the far face could thus be obtained, its slope being taken as a measure of the thermal diffusivity (Table 1).

Table 1. *Rise in temperature of compacts*

	Thickness of compact mm	Rise in temperature of compact face $^{\circ}\text{C s}^{-1}$
Asagran .. .. .	4.56	0.052
Boric acid .. .. .	4.69	0.176
Sodium chloride .. .. .	4.76	0.41

*Materials*

The substances chosen for investigation were Asagran, a proprietary form of acetylsalicylic acid, Monsanto Chemicals Ltd., sodium chloride and boric acid both as crystalline B.P. material. These were not treated further except that some samples were prepared by shaking the materials with 2% w/w of magnesium stearate for use as a lubricant.

## RESULTS AND DISCUSSION

*Interpretation of the traces*

Asagran (Fig. 2A) shows an initial temperature rise on compression (a-c) followed by a slow fall (c, d) as heat leaks out into the punch and die. To keep the trace within a reasonable length, the Visicorder was stopped at intervals until the spot returned to the base line. The true temperature rise was found by graphical extrapolation of the cooling curve to cut a perpendicular erected midway through the period of observation.

Release of the compression force after the compact had returned to room temperature led to in every case a rapid fall in temperature (e.g. d, e in fig. 2A) followed by a slow rise as heat leaked back into the compact. On recompression to a force of 50 kN, a rise in temperature again occurred but to a value lower than the initial rise. On relaxation a fall to a value similar to that of the first relaxation was noted. The temperature rise and fall on recompression and relaxation could be reproduced indefinitely and the process would seem to be analogous to the adiabatic compression and expansion of a gas.

We would suggest that the temperature rise on initial compression can be separated into two components: (1) that associated with the heat of formation of the compact and probably due to granule friction, plastic deformation and bonding; (2) that associated with the storage of strain energy within the formed compact, energy which is largely recovered when the compression force is removed. If the compact has cooled to room temperature then a fall in temperature results when the compact relaxes. The slope of the heating curve on initial compression changes significantly at b (Fig. 2A)

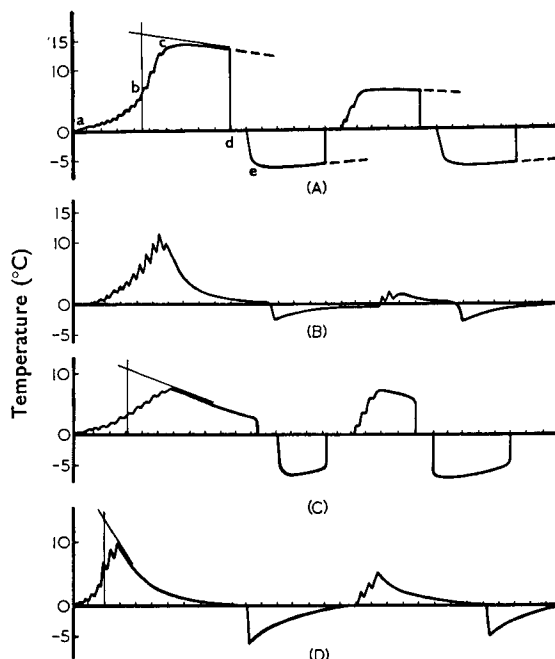


FIG. 2. Oscillograph traces resulting on compression, recompression and relaxation of unlubricated compacts (approximately half actual size). Compression force 50 kN. Time scale in s. (A) Asagran 2.0 g., (B) Sodium chloride 2.0 g., (C) Boric acid 2.0 g., (D) Asagran 0.5 g.

although pressure was applied at a constant rate. This rise can be attributed to compact formation while the further rise b—c is due to its compression.

The trace for sodium chloride (Fig. 2B) shows the same effects but because the thermal diffusivity of its compact is much greater than that of an Asagran compact (Table 1), there is a very rapid fall to room temperature. Although the slope is too steep for graphical extrapolation as used in Fig. 2A to be used here, the rise in temperature would appear to be comparable with the results obtained by Hanus & King (1968). It is likely that the temperature rise on recompression is related to the strain energy per unit weight stored in the compact, the low temperature rise on recompression of sodium chloride would then indicate a correspondingly low value for this quantity. It is known (Ridgway, Glasby & Rosser, 1969) that sodium chloride crystals have a high value of Young's modulus while that for aspirin crystals is low. Provided the compacts have similar properties, a sodium chloride compact would thus store less energy than one of aspirin when compressed with the same force.

Boric acid, sometimes used as a lubricant for soluble tablets, has crystals which are slippery. The strain energy contribution to the temperature rise on initial compression is a large part of the whole temperature rise and the rise due to compact formation is only about 2° C (Fig. 2C). The low interparticulate friction probably accounts for the low temperature of formation.

#### *Temperature rise or fall on recompression or relaxation*

These are plotted against the compression force for all three substances in Fig. 3A and B. The results support the observation of Hanus & King (1968) that temperature rise is directly proportional to compression force. Since these authors used calori-

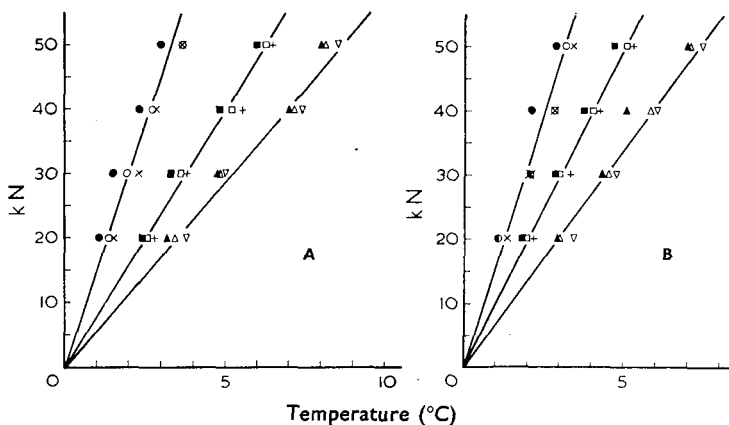


FIG. 3. A. Temperature rise attained on recompression v. compression force. B. Temperature fall on relaxation v. compression force. All compacts were unlubricated. Measurements were made on three separate compacts. Compact weight was always 2.0 g. Sodium chloride: ●○× Asagran: ■□+. Boric acid: ▲△▽

metric measurements on ejected tablets, they obtained the net temperature rise on formation and not the rises on compression or the fall on relaxation.

For boric acid and Asagran compacts the temperature fall on relaxation is always less than the rise on recompression indicating that only a proportion of the strain energy is recovered. We would predict that the compression force versus punch displacement curves for both these substances would show pronounced hysteresis loops representing the energy which is not recovered on relaxation. The high thermal diffusivity and small temperature changes of sodium chloride compacts make difficult any conclusion about the magnitude of the temperature rise and fall on its recompression and relaxation.

*The effect of compact size and lubrication*

The effect of varying the weight of the compacts is shown in Table 2. Although some differences in the temperatures of formation, recompression and relaxation occur, these are not pronounced and can be attributed to experimental and extrapolation

Table 2. Temperature changes °C in Asagran compacts of varying weight compressed at 50 kN

Compact weight (g)	Temperature rise on initial formation	Temperature fall on first relaxation	Net rise in temperature (attributed to heat of formation)	Temperature rise on recompression	Temperature fall on relaxation
0.5 (U)	15.0	6.2	8.8	7.4	5.7
1.0 (U)	15.8	5.6	10.2	7.4	5.7
1.5 (U)	14.4	5.2	9.2	6.1	5.1
2.0 (U)	*15.8	*5.6	*10.2	*6.2	*5.0
0.5 (L)	14.1	5.1	9.0	7.4	5.9
1.0 (L)	14.1	6.8	7.3	8.5	6.6
1.5 (L)	13.4	5.2	8.2	6.8	5.7
2.0 (L)	13.4	5.1	8.3	6.4	5.7

(U) Unlubricated compact

(L) Compact lubricated with 2% w/w magnesium stearate

\* Mean of three results on three separate compacts.

errors. The temperatures attained are apparently independent of the compact weight and this is consistent with the idea that it is the energy supplied or given out per unit weight that determines their magnitude.

Juslin (1969) found that 1% w/w of calcium stearate greatly reduced the temperature rises on the surfaces of compressed tablets caused by die wall friction on ejection. In contrast, the addition of 2% w/w magnesium stearate to Asagran granules does not appreciably affect the internal temperature rises (Table 2). Higuchi, Rao & others (1953) and Shotton & Ganderton (1960) have shown that tablet formation is accompanied by granule fracture and rebonding. Lubricants, when present at their usual low concentrations, may have limited effect on the heat produced during these processes.

Where the material itself has lubricant properties, as with boric acid, then the much larger lubricating effect of the material could be effective during granule fracture and rebonding and may account for the low net temperature rise on formation of a compact of boric acid.

#### *Acknowledgements*

We should like to thank Mr. L. Tibbs, Chief Technician in The School of Engineering for helpful technical advice and the extended loan of the Visicorder, and Mrs. R. Faulkner for her skilful preparation of the illustrations. We are also indebted to Dr. B. G. Knowles for valuable discussion during the course of the work.

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