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The compaction properties of sodium chloride in the presence of moisture

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The effect of moisture on the behaviour of sodium chloride under compression was investigated by measurement of compaction forces and ejection forces. Saturated solution present at the crystal surfaces effected a reduction in friction at the interparticulate boundary and between the particles and the die wall. Dry material formed compacts of higher strength than wet material except at low pressures. Subsequent drying of compacts prepared in the presence of moisture resulted in an increase in strength due to interparticulate recrystallisation.

MANY of the difficulties encountered during the preparation of compressed tablets may be attributed to an unsuitable moisture content. The effects of moisture on the compaction properties of peat (Matveev, 1951; Naumovich, 1957), coal (Crone & McKee, 1950), soil (Maclean, 1948; Lewis, 1959) and brick (Boyd, 1949) have been reported. The present work was performed to investigate the influence of moisture on the behaviour of a soluble particulate solid during compression, and on the properties of the compact produced. Jaffe & Foss (1959) state that removal of water of crystallisation prevents the formation of tablets from materials which normally bond by direct compression. Other reports (Ferrand, 1955; Martin & Cook, 1961) have quoted values of relative humidity at which the preparation of compressed tablets is facilitated. Discrepancies between these values suggest that optimum ambient conditions are specific to individual materials. Seth & Münzel (1959) and Egorova & Vikul'eva (1961) derived an optimum granular moisture content which was specific to a given granulation and yielded tablets of greater strength. At higher moisture content adhesion of material to the punch faces occurred whereas, in certain cases, a low moisture content resulted in "capping" and lamination of the tablets.

In the present investigation it was considered advisable to avoid additives and sodium chloride was consequently selected as an ionic, cubic crystalline material, capable of being compressed directly to form a coherent compact.

Experimental

A Lehman single punch eccentric tablet machine was instrumented with Saunders Roe $\frac{1}{8}$ inch linear foil resistance strain gauges (60 ohm nominal resistance, gauge factor 2·07) in a manner similar to that described by Shotton & Ganderton (1960). Two gauges were bonded to the 1·2 cm diameter plane-faced upper punch (K9 tool steel, surface ground to 5 microinches) using Eastman 910 cement, and three gauges were bonded to the lower punch holder. The gauges and terminals were then coated with a polysulphide rubber compound. The technique of Shotton & Ganderton (1960) was employed to calibrate the strain gauge circuits. The

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machine was installed in a room in which the ambient relative humidity was controlled.

The weighing assembly of a Cahn Gram Electrobalance was installed in a vacuum oven and employed for thermogravimetric determination of moisture. A loss in weight of 0·05 mg could be detected in a sample weight of 900 mg (equivalent to approximately 0·005% moisture).

A batch of sodium chloride (B.P. quality) was screened to obtain a 30–40 mesh fraction. This fraction was then subjected to further separation on a Lavino Alpine Air-Jet Sieve to remove fines of −100 mesh size adhering to the surface of the larger crystals.

Weighed samples of the screened sodium chloride, sufficient to produce a compact of 0·4 cm thickness at zero porosity in a 1·2 cm diameter die, were stored in glass vials contained in a constant humidity chamber. The time of storage depended on the number of samples present and was varied to produce a range of moisture contents. When this had reached the required value the containers were removed from the humidity chamber and rapidly sealed. Preliminary experiments with the Cahn balance had indicated a high rate of moisture loss from the surface of cubic crystals at normal ambient conditions. Consequently, the conditions in the room were maintained identical to those in the humidity chamber.

Six humidified samples were compressed at each selected pressure and measurements of upper and lower punch forces and ejection forces were recorded. The resulting compacts were weighed and the dimensions of each were measured. Three compacts at each pressure level were tested using a diametral crushing apparatus (Shotton & Ganderton, 1960). The three remaining compacts were similarly examined after drying to constant weight over silica gel.

A binocular microscope was employed to examine the fractured surfaces of the compacts.

The compressional behaviour of sodium chloride, previously dried at 110° and in which no moisture could be detected, was investigated by two methods. In one experiment, the die wall was "conditioned" by compaction of two samples of dry material before measurements were made of compression forces on subsequent samples. The die was cleaned before compression of samples at a different pressure. In the second experiment the die was cleaned before the preparation of every compact.

Results

Compression

The effect of moisture on the proportion of force transmitted to the lower punch through the mass is shown in Fig. 1, where R is the ratio of maximum transmitted force, F_B, to maximum applied force, F_A, during a compression cycle. A low moisture content (0·02% w/w) produced a large increase in F_B at low applied force compared with anhydrous sodium chloride compressed in a conditioned die. As the applied force increased, the punch force ratio decreased.

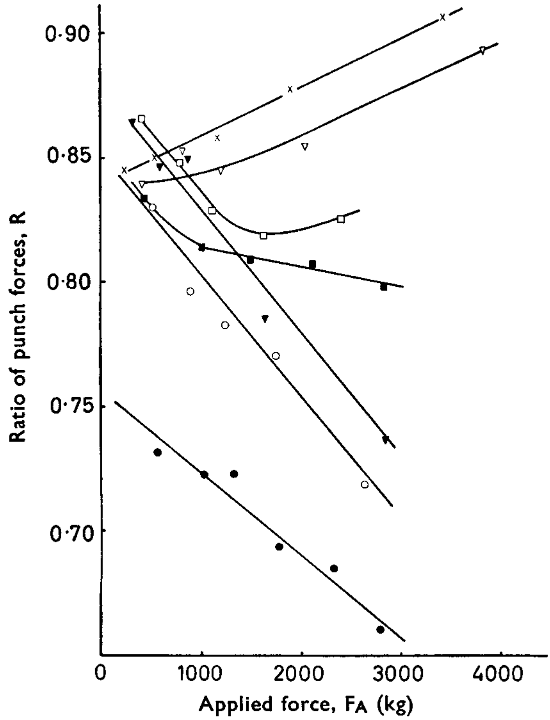

FIG. 1. Effect of moisture content on the ratio of transmitted punch force to applied punch force. Moisture content %: ×, 10; ▽, 2·4; □, 0·55; ■, 0·16; ○, 0·02; ▼, 0 (clean die); ●, 0 (conditioned die).

Force lost to the die wall, F_D, due to frictional resistance is equivalent to the difference in the values of applied and transmitted force, and variation of this parameter with increasing applied force is recorded in Fig. 2. For 0·55% moisture, the relationship was linear up to 1600 kg applied force (Fig. 2); above this the slope of the graph decreased and the punch force ratio, R, increased (Fig. 1). In the presence of 2·4 and 10% moisture, the values of R at low applied force were lower than for 0·55% moisture, and a continual increase in R occurred with increasing applied force. At high values of applied force, an increase in moisture content resulted in an increase in transmitted force.

EJECTION

Anhydrous sodium chloride compressed in a conditioned die exhibited a linear relationship between ejection force and applied force (Fig. 3). A reduction in ejection force was obtained when such samples were compressed in a clean die.

At all values of applied force greater than 700 kg, ejection was facilitated by an increase in moisture content. Above 1700 kg, applied force, the values of ejection force were higher than expected for moisture contents of less than 0·55%. Conversely, ejection forces obtained for 2·4% and 10% moisture content were lower than anticipated by extrapolation of the linear portion of the graphs.

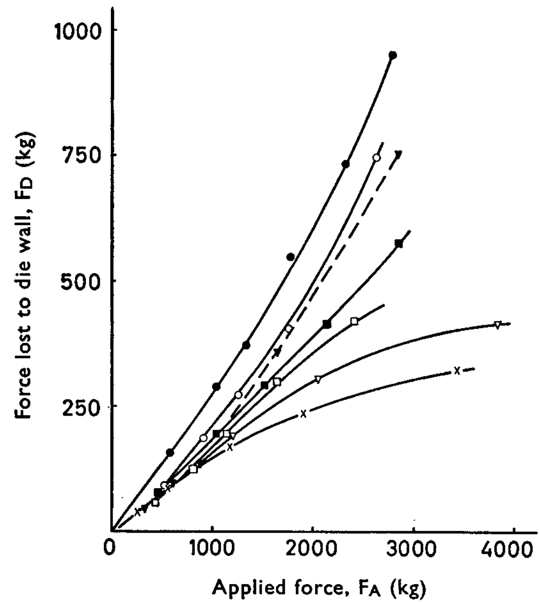

FIG. 2. Effect of moisture on the relationship between applied force and force lost to die wall. Key as in Fig. 1.

For anhydrous sodium chloride, the relationship between the force lost to the die wall and the ejection force at a given applied force was independent of the condition of the die wall (Fig. 4). Moisture exerted a greater effect at the die wall during ejection than during compression, and deviation from the initial relationship for dry material occurred at values of F_E which depended on the moisture content. Above these values of F_E, a common relationship between F_D and F_E was obtained for all samples containing moisture.

CRUSHING STRENGTH

Compacts of anhydrous sodium chloride compressed in a clean die were stronger than those prepared in a conditioned die (Fig. 5). Increases in strength at low pressures were obtained in the presence of 0·02% and 0·55% moisture, compared with anhydrous samples compressed in a conditioned die. Samples containing 10% moisture produced compacts of lower wet strength than 0·55% moisture at all pressures. At high pressures, all samples containing moisture produced compacts of lower strength than the anhydrous material.

The desiccation of moist compacts produced a strength increase which was proportional to the amount of moisture remaining after compression.

During the compression of samples containing 10% moisture, saturated solution of sodium chloride was expelled from the die and at a mean applied pressure (P_M) of 3000 kg/cm² only 3% moisture remained in the compact. The percentage increase in strength on drying of compacts compressed in the presence of 10% original moisture was thus greater at low pressures.

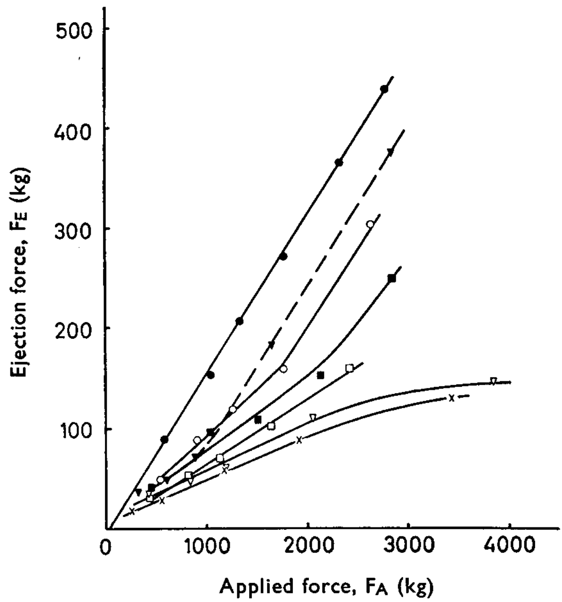

FIG. 3. Effect of moisture on the force required for ejection. Key as in Fig. 1.

Discussion

Conflicting results have been reported concerning the relative importance of the frictional effects at the interparticulate boundary and the particle-die wall interface (Bal'shin, 1938; Seelig & Wulff, 1946; Kamm, Steinberg & Wulff, 1947; Torkar, 1956). Carrington (1958) found an increase in force lost to the die wall when radial movement of particles towards the die wall was facilitated by interparticulate lubrication or punch face lubrication, or both. When lubricant was present at *all* interfacial junctions, a decrease in die wall reaction compared with an unlubricated system indicated that the die wall effect was greater than the interparticulate effects.

The increase in punch force ratio in the presence of 0·55% moisture at low applied force (Fig. 1) may be explained by a reduction in friction due to a lubricant moisture film at the die wall. The force lost to the die wall increases with applied force as the area of compact in contact with the die wall increases. As the porosity of the compact decreases, the void spaces become filled with liquid. Increases in applied force then cause expulsion of liquid to form a continuous film at the die wall. The liquid reduces the coefficient of friction between particles and die-wall, and also restricts movement of solid into contact with the die. Consequently, for a given increase in applied force there is a smaller increase in contact area, A, between compact and die wall. Assuming that F_D is proportional to A, an increase in applied force then produces only a small increase in F_D.

Lower moisture contents (0·16% and 0·02%) provided less die-wall lubrication at all values of applied forces. No migration of liquid to the die wall occurred even at high applied force, since sufficient void space remained to accommodate the small volume of liquid.

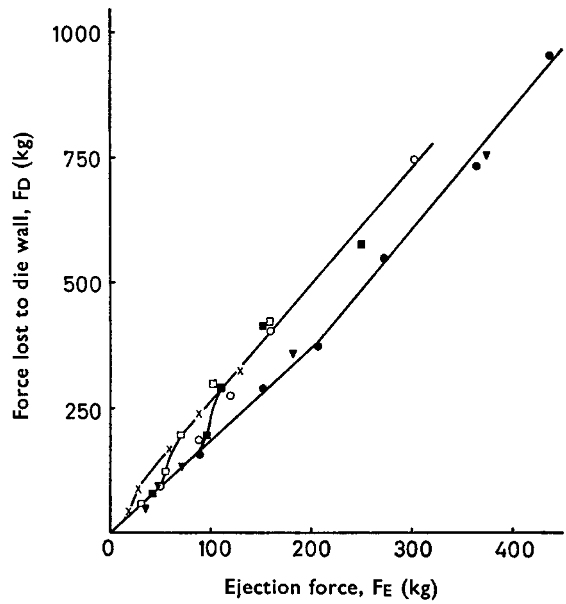

Fig. 4. Effect of moisture on the relationship between the force lost to the die wall and the ejection force. Key as in Fig. 1.

For the theoretical case when all air is preferentially eliminated from the sample containing 10% moisture, it was estimated that when the porosity decreases to 20%, saturated solution will fill the void spaces. In this condition the application of small loads may produce a lateral displacement, or flow, of particles into contact with the die, to yield a higher die wall reaction than for 0·55% moisture (cf. Carrington, 1958). As the applied force increases, liquid is expelled from within the compact and die wall lubrication increases continuously.

The lower values of die wall friction and ejection force for a "clean" die compared with a die which had been "conditioned" confirm that successive compressions increase the extent of die-wall contamination by compressed material. Contamination of the die wall occurs when the shear strength of the bond between the steel die and material becomes greater than that of the material itself. Stephenson (1965) observed a cyclical contamination of the die wall, and a resulting increase in die wall friction, during compression of anhydrous potassium bromide granules. An increase in moisture content of the granules to 0·5% abolished this effect.

Microscopic examination showed that the presence of water in the sodium chloride caused a rounding off at the edges and corners of the initially cubic crystals, owing to preferential dissolution in these localities. The lubricant effect of saturated sodium chloride solution will be greater between plane or smooth surfaces, whereas points and edges will penetrate the film. The change in crystal shape may thus facilitate movement in the bed and reduction in the incidence of shear failure of crystals.

Increased consolidation of the compact permitted by a lower frictional resistance at the die wall explains the increases in strength of compacts produced by compression in a clean die and in the presence of moisture at low pressures.

A small increase in strength in the presence of moisture may be produced by a surface tension effect which assists the adhesion between particles. This effect will be relatively large where the interparticulate bond is weak and when the number of bonds is small.

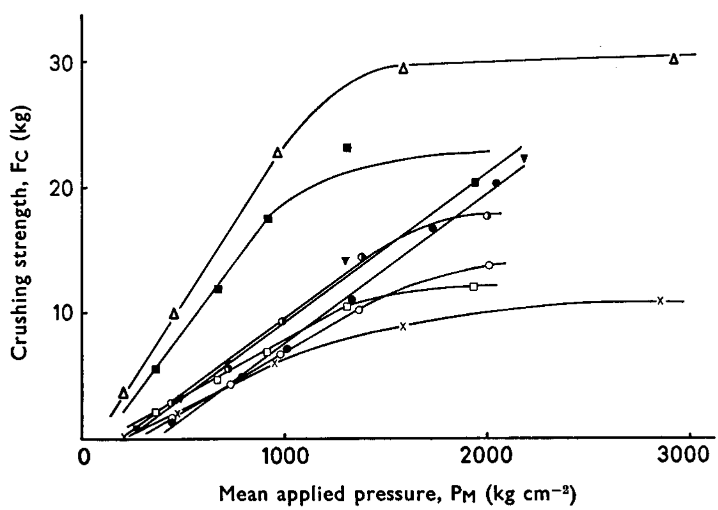

FIG. 5. Effect of moisture on the crushing strength. Moisture %: △, 10 (dried), ×, 10 (wet); ■, 0·55 (dried); □, 0·55 (wet); ◑, 0·02 (dried); ○, 0·02 (wet); ▼, 0 (clean die); ●, 0 (conditioned die).

It appears that 10% moisture exerted a hydrodynamic resistance to consolidation which counteracted the lubricant effects. Despite the low viscosity of the liquid film, lubrication inhibited interparticulate shear forces and thus reduced the amount of bonding which occurred at high pressures. This is in agreement with the results of Strickland, Nelson, Busse & Higuchi (1956) and Lewis (1964) for solid lubricants. Subsequent examination of the fractured surface revealed that failure had almost entirely occurred around the particles (Shotton & Ganderton, 1961) in compacts prepared at low pressure from samples with 10% moisture. At higher pressures, expression of liquid from the compact permitted interparticulate contact, and evidence of fracture across the crystals increased. Decreasing moisture content resulted in a greater incidence of cross-grain fracture associated with an increase in strength. Anhydrous sodium chloride, compressed at high pressures, showed only cross-grain fracture.

Microscopic examination of the dried compacts confirmed that the increases in strength are mainly due to recrystallisation at the crystal boundaries and in the void spaces. Surface tension forces will produce a tendency for the solution to flow into positions where crystallisation will produce a maximum effect, i.e. at the angles of contact between crystals. Crystallisation of the saturated solution occurring at discontinuities on the crystal surface will effect an increase in crushing strength because it is at such faults that propagation of fracture usually originates (Griffith, 1920).

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