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Granulation and compaction of a model system. II. Stress relaxation

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Introduction

A previous paper (Cutt et al. 1986) has described the physical characteristics of granules prepared from a model material, glass ballotini, by massing and screening. Wet granulation using aqueous solutions of binders may alter the compression properties of the substrate, in particular the ability to deform plastically. This effect may be studied by measuring the decay of axial stress with time, the stress relaxation of the material. This paper examines the stress relaxation characteristics of glass ballotini granules prepared with different binders.

Materials and Methods

Granules were prepared by wet granulation from lead glass ballotini (Dragonit 30, Grade 20, Englass Ltd., Leicester, U.K.), fractionated to give a mean size of 40 μm (standard deviation 8 μm). The granulating agents used were polyvinylpyrrolidone (PVP) average mol. wt. 25,000 Da (Kollidon 25, BASF, Ludwigshafen F.R.G.,); hydroxypropylmethylcellulose (HPMC) low viscosity grade (Methocel E15, Colorcon, Orpington, U.K.); hydrolysed gelatin, average mol. wt. 10–12,000 Da (Byco C, Croda Foods, Widnes, U.K.). Full details of the procedures have been described previously (Cutt et al., 1986). Stress relaxation measurements were made using an Instron Physical Testing Instrument modified to take a conventional 1.27-cm diameter flat-faced punch and die system (Fell and Newton, 1970). Weights of material calculated to occupy a volume of 0.3 cm^3 at zero porosity were compressed at 0.2 cm/min in a prelubricated die to the required axial force. The cross head was then halted and the decay of axial force recorded over a period of 6 min. Corrections for relaxation of the instrument were performed by a similar procedure with no material in the die.

Results and Discussion

The data obtained from the stress relaxation of the glass and granules in this study did not show complete stress relaxation even after 6 min, nor did the materials exhibit the characteristics of a true Maxwell body. Hence it was not possible to represent the decay in axial stress by a linear viscoelastic model. Log–time plots were therefore chosen to illustrate the stress relaxation data, as the expansion of the time scale immediately after the peak force allowed comparison of the materials during the early stages of relaxation. The physical significance of changes in slope is unknown, but brittle materials exhibit a slower and less total relaxation compared with plastic materials (Hiestand et al., 1977).

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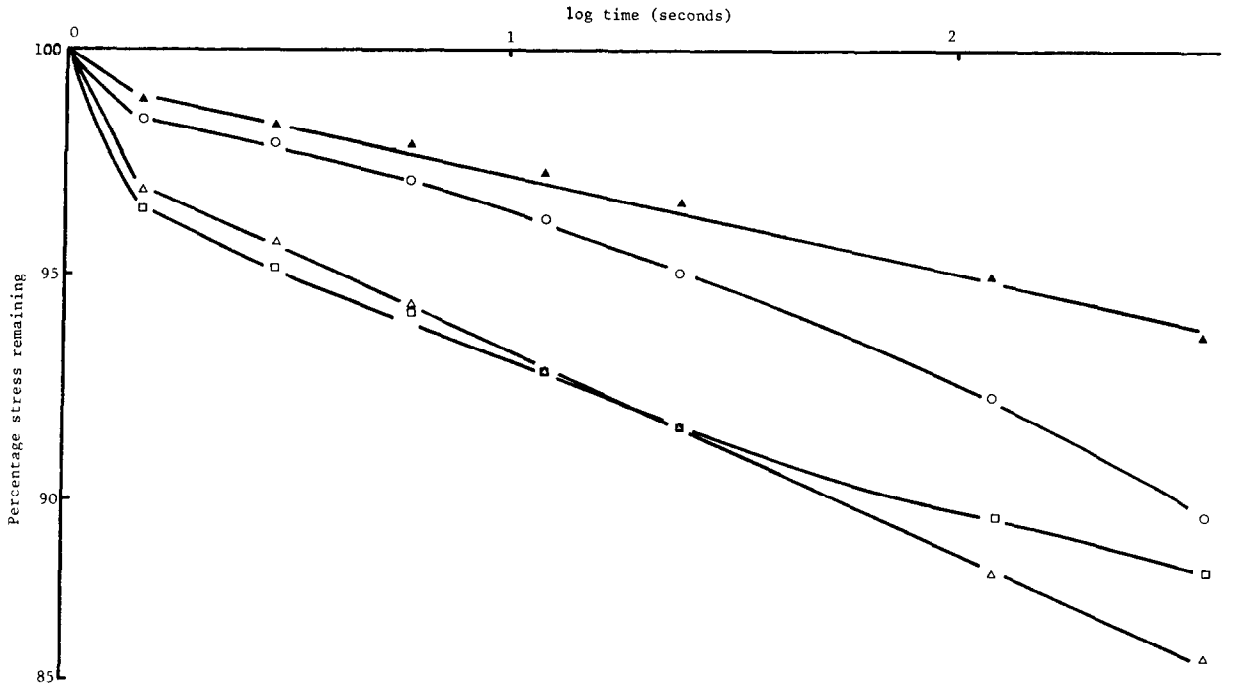

Fig. 1. The effect of granulation and binder type on the stress relaxation of ungranulated and granulated glass beads using 1.5 % w/w binder. ▲, No binder; ○, PVP; □, Byco; △, HPMC. Storage equilibrium relative humidity, 12%; maximum applied pressure, 39 MN/m^2.

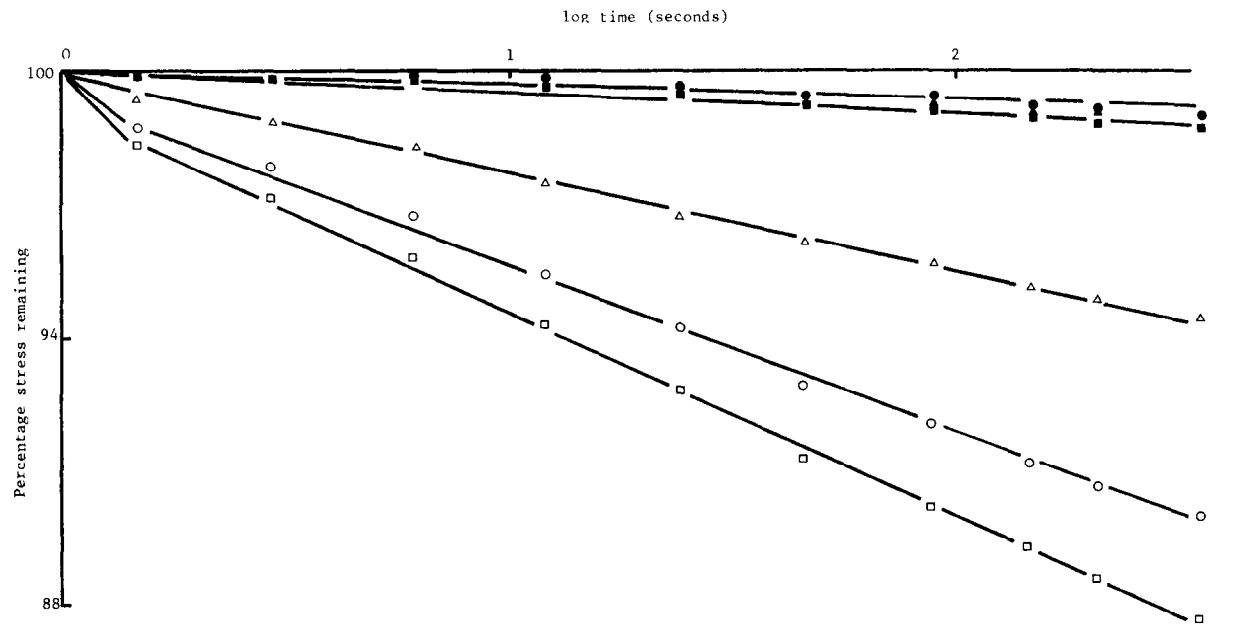

Fig. 2. The effect of the moisture content on the stress relaxation of compressed binders. Applied pressure, 194 MN/m^2. △, 0% w/w HPMC; ▲, 14.5% w/w HPMC; ○, 0% w/w PVP; ●, 14.3% w/w PVP; □, 0% w/w Byco; ■, 14.9% w/w Byco.

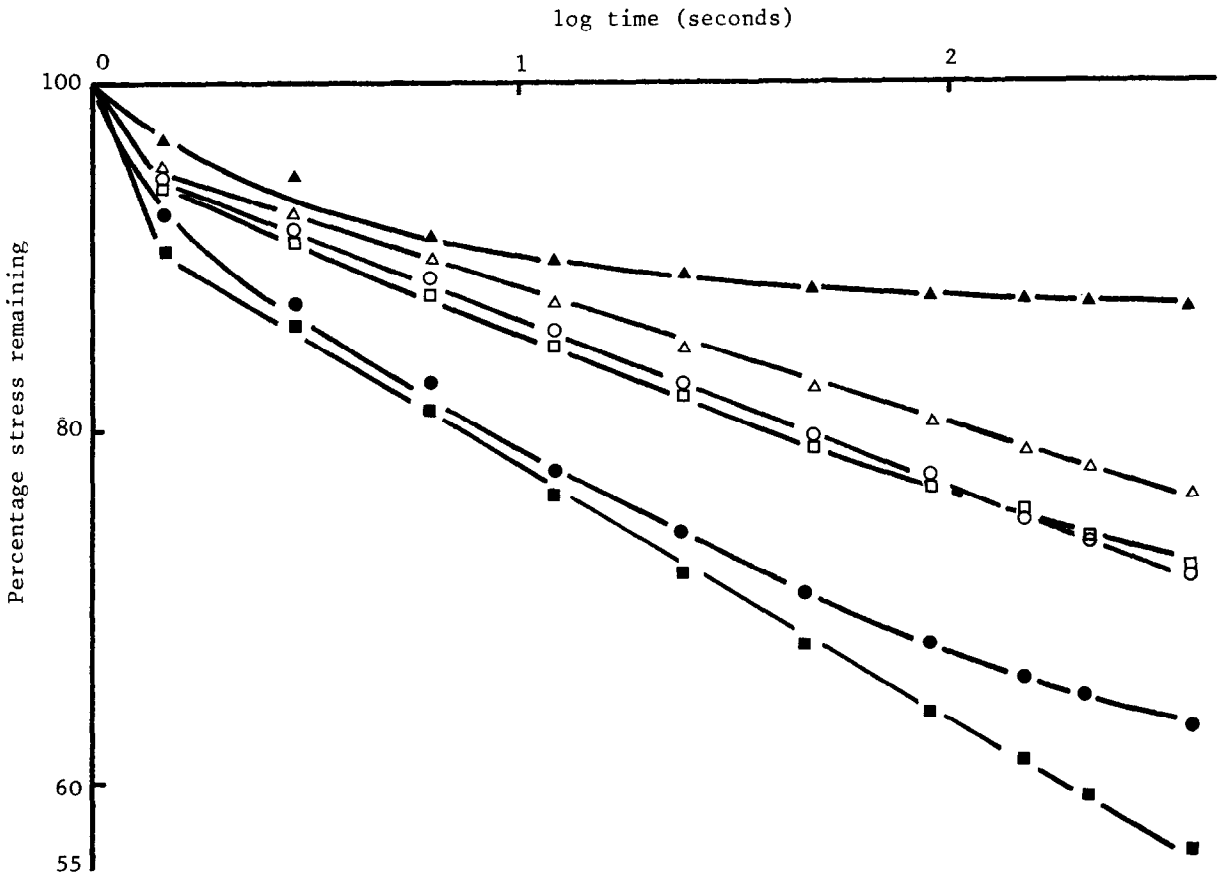

Fig. 3. The effect of moisture content on the stress relaxation of compressed binders. Applied pressure, 39 MN/m^2. Key as in Fig. 2.

The relaxation of the glass beads alone is illustrated in Fig. 1, where the results are compared to granules prepared from the beads. Some relaxation of the glass alone is noted and as the glass would not deform plastically under the tabletting conditions used, the relaxation was attributed to further repacking of the ballotini or to fracture by a static fatigue failure mechanism (Walton, 1958) or a combination of the two.

The stress relaxation behaviour of the compacted binders alone is shown on Figs. 2 and 3. The rank order of stress relaxation at both pressures and moisture contents was Byco > PVP > HPMC. At the higher pressure, porosity measurements on the damp binders indicated that they had almost totally deformed leaving few voids into which relaxation could occur. The differences betweeen materials can therefore be illustrated more clearly by the use of a maximum axial load that is great enough to induce plastic deformation, but not so large as to plastically deform the material totally during the compression phase. The rank order of stress relaxation may be related to the particulate characteristics of the materials. Both Byco and PVP consist of small hollow spherical particles, whereas the particles of HPMC are larger and denser.

All the granules tested exhibited a significantly faster and greater total relaxation during the experimental time than the glass powder alone, as shown on Fig. 1. The glass beads in granules containing 1.5% w/w Byco theoretically could have a 0.22 μm thick coat of this binder, if it were spread uniformly over their surfaces. This coating would be approximately 0.5% of the mean particle diameter of the glass. Nevertheless, the granules

TABLE 1

The effects of binder type and concentration, equilibrium storage humidity and maximum force on the stress relaxation of glass granules

		PVP			HPMC			Byco		
Binder concentration (% w/w)										
NOMINAL		1.5	3.0	4.5	1.5	3.0	4.5	1.5	3.0	4.5
ASSAY		1.65	3.26	4.83	2.09	3.08	4.45	1.41	3.02	4.50
Stress loss (%)										
39 MN/m^2	12% R.H.	10.4	14.6	16.6	13.6	16.0	14.8	11.8	14.4	15.0
	65% R.H.	12.6	15.2	18.8	14.0	16.2	16.0	16.0	20.4	22.8
194 MN/m^2	12% R.H.	6.0	5.9	6.4	6.8	7.2	6.9	6.0	5.9	6.2
	65% R.H.	8.3	8.8	9.2	7.6	8.0	7.8	7.0	7.6	7.8

Stress loss after 6 min. Initial strain rate 0.2 cm/min; granule diameter 710–1000 μm. R.H., equilibrium relative humidity.

containing this low concentration of binder were able to exhibit an increased stress relaxation compared with the glass alone. The porosity of the majority of the granules under pressure was lower than that of the glass alone. Hence, porosity differences were not responsible for the increased stress relaxation of the granules. The binder itself may deform plastically in order to relieve the stress at points of contact with the glass or compression tooling. The binder may also ease the deformation of the granule, under load, into the remaining voids, and hence reduce the axial stress on the compact.

The stress relaxation behaviour exhibited by similar granules which varied only in the binder used did not show a consistent trend. Table 1 shows that at the lower compression force and higher storage humidity, Byco granules relaxed more than the HPMC or PVP granules at each of the 3 binder concentrations studied. However, at the higher compression force, the PVP granules relaxed more than the other damp granules. The relaxation behaviour of the drier granules, stored at 12% relative humidity, was different again. The HPMC granules generally relaxed the most, irrespective of the compression pressure, except at the lower force and highest binder concentration when PVP was ranked first. The porosity of these granules under compression was dependent on the binder type. These differences in compact porosity, and also the bonding and distribution characteristics of each binder, may produce the different rank orders for granule relaxation profiles with changes in the compression pressure. Hence, relaxation studies on the binders alone do not give a good guide to the behaviour of the granules.

No significant difference ($P = 0.95$) was found between the stress relaxation behaviour of small (125–250 μm) and large (710–1000 μm) granules (Table 2). Shlanta and Milosovitch (1964) showed

TABLE 2

The effect of granule size on the stress relaxation of glass granules containing 3% w/w binder

		HPMC		PVP		Byco	
		12% R.H.	65% R.H.	12% R.H.	65% R.H.	12% R.H.	65% R.H.
125–250 μm granules	39 MN/m^2	85.0	84.6	86.2	84.2	85.6	78.8
	194 MN/m^2	92.6	91.9	94.2	92.2	94.4	91.4
710–1000 μm granules	39 MN/m^2	84.0	83.8	85.4	84.8	85.6	79.6
	194 MN/m^2	92.8	92.0	94.1	91.2	94.1	92.4

Experimental values remaining stress (%) after 6 min.

that particle size was an important factor in the stress relaxation of powders, but in the current study, the inherent weakness of the granules would give rise to breakdown to constituent glass beads and deformed binder at low pressures.

An increase in binder concentration resulted generally in an increase in both the rate and magnitude of stress relaxation (Table 1). This would be a reflection of the thicker coating of material surrounding the beads, and an increase in the quantity of binder in each bond. Experiments with hydrophobic glass, prepared from the beads by treatment with dimethylsilane (Mohammad and Fell, 1982) showed no significant differences in the stress relaxation behaviour of granules prepared for this material to that of the hydrophilic glass.

The use of a non-plastically deforming model material has shown that granulation using binders introduces a significant plasticity to the resultant granules. Comparison between the stress relaxation of the binders alone and the granules prepared from the binders is inconclusive, but increases in moisture content and binder concentration markedly change the plasticity of the granules.

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