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Effect of fluid viscosity on the size and morphic features of salicylic acid during ball-mill grinding

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Summary

The effect of ball milling on the morphic features of salicylic acid crystals was studied in wet and dry grinding. Fourier shape descriptors of individual particles were computed using an image analysis system. Data obtained indicated that the ball milled particles differ significantly from the original crystals with regard to size and shape parameters. The degree of roundness and elongation of the fragments obtained depend on the viscosity of the grinding media.

Introduction

The importance of particle shape in determining the physical properties is now recognized in a great variety of industries (Meloy, 1984; Beddow and Meloy, 1980; Beddow et al., 1980; Laurin, 1985). In a recent communication from our laboratory it was found that the morphological characteristics of talc powder can significantly affect the physical behaviour of this excipient as an opacifier (Laurin, 1986). The change in the morphological parameters of salbutamol solid particles was also studied after jet mill grinding (Akbarieh and Tawashi, 1987). Results obtained indicated that microgrinding produced particles with smoother boundaries, less elongation and a higher degree of roundness (Akbarieh and Tawashi, 1987).

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Although milling and micronization have been used successfully in the production of powders for granulations, suspensions and aerosol-formulations, only few studies are published on the shape of the particles produced after size reduction (Lantz, 1982; Barnett and Sims, 1983). There is also no experimental evidence on the relative importance of the material properties, as compared with the method of breakage, in determining the particle shape (Holt, 1980). The objective of this paper is to examine the effect of dry vs wet grinding on particle shape and to study the importance of fluid viscosity on the morphic features during size reduction.

Materials and Methods

Throughout this study, salicylic acid crystals, 60–120 mesh (Sigma, St. Louis, U.S.A.) were used (see Fig. 1).

As grinding media, three liquids of different viscosities namely kerosine (Fisher, Montreal,

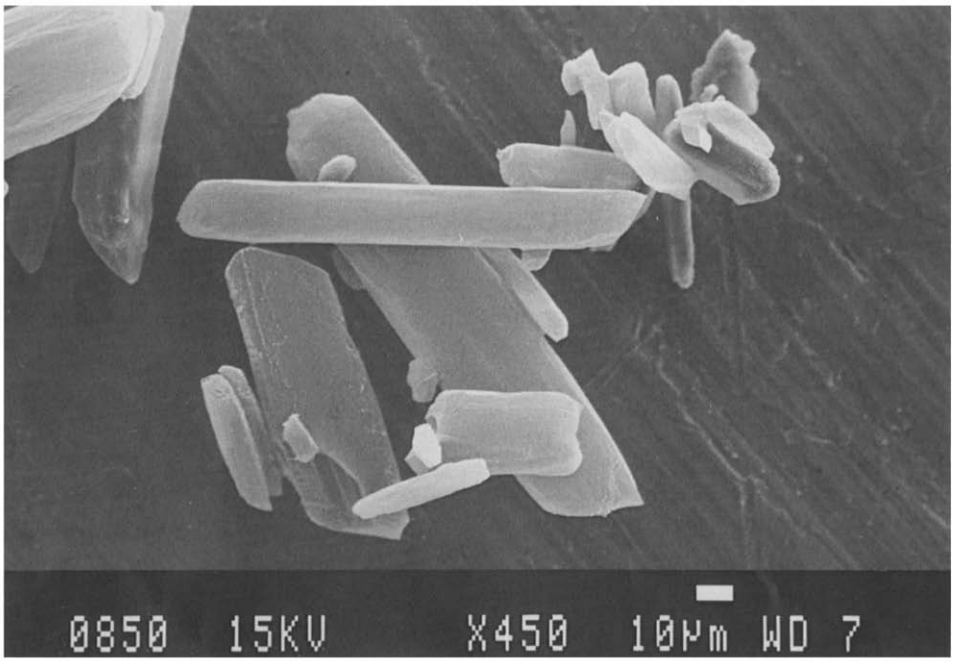

Fig. 1. Scanning electron micrograph of salicylic acid crystals.

Canada), light liquid paraffin (LLP) and heavy liquid paraffin (HLP), (Sigma, St. Louis, U.S.A.) were used. Viscosity was measured by a rotary viscometer (Haake Rotovisco, Berlin-Lichterfelde, F.R.G.).

A ball mill of porcelain jar and porcelain balls (Erweka GmbH, F.R.G.) was used at 27 rpm. The volume of the balls was 650 ml. Each charge consisted of 40 g of salicylic acid crystals, to which 200 ml fluid was added. The dimensions of the mill are: diameter = 19.27 cm, volume = 4750 ml, height = 16.30 cm.

Size and shape analysis of salicylic acid crystals before and after grinding were performed by an image analysis system previously described (Akbarieh et al., 1987). The system is based on the digitization of the particle image by obtaining (x, y) coordinates of the particle boundary (Granlund, 1972, Chen and Chi, 1981; Nguyen et al., 1983). Size and shape features characterizing the particle such as diameter, shape spectra, roundness and the degree of elongation were determined as previously reported (Dubuc et al., 1987 and Akbarieh and Tawashi, 1987). At least 100 crystals were analysed for each time interval during wet and dry grinding.

Results and Discussion

The results of size reduction (Fig. 2) show the effect of ball mill grinding on particle size in the

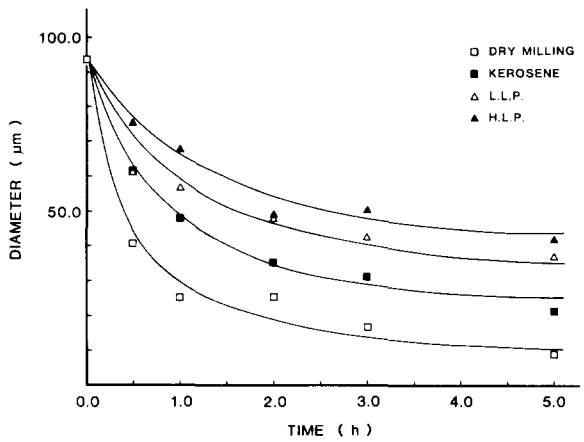

Fig. 2. Effect of grinding on particle size as a function of time.

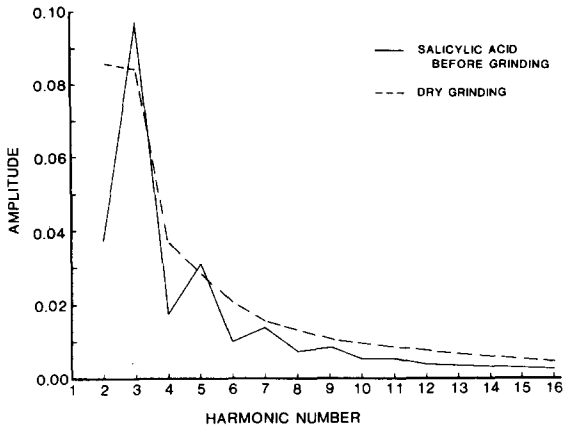

Fig. 3. The amplitude vs harmonic number for salicylic acid crystals before and after dry grinding.

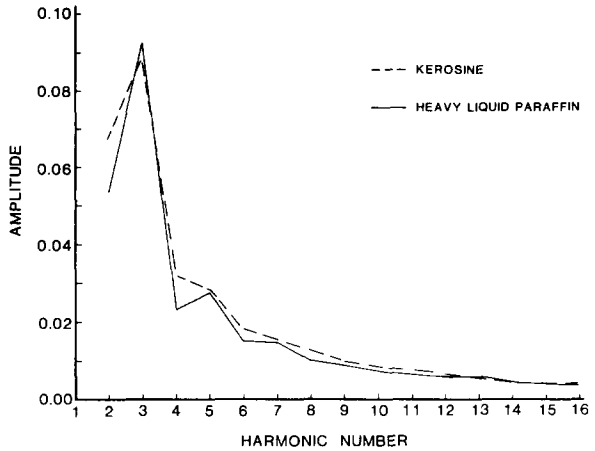

Fig. 4. The amplitude vs harmonic number for salicylic acid crystals after wet milling using kerosine and HLP as grinding media.

dry and wet conditions. Dry grinding produced particles smaller in size than wet grinding. In presence of kerosine, the size reduction process was faster than in LLP and HLP. The presence of a fluid as a grinding medium altered to a great extent the rate of size reduction. Although it has been reported (Perry, 1963; Prem, 1984) that finer sizes could be achieved by wet than by dry milling for certain solids, the results obtained in these experiments indicated the opposite. This can be explained by the fact that the effective forces acting on the particles in the ball mill have been significantly reduced because of the viscosity of the liquid vehicle. This in addition to the change in the location of the suspended particles and partial agglomeration. The combined effects of these factors on the force used in size reduction resulted in plastic deformation rather than fragmentation of test material.

Figs. 3 and 4 show the effect of grinding on the particle shape in the dry and wet conditions. The amplitude vs the harmonic number indicated a clear morphological difference between the particle subjected to dry and wet grinding. The effect of fluid viscosity on the morphological parameters, namely particle roundness and particle elongation ($\pm$S.E.M.) is given in Figs. 5 and 6. The degree of roundness decreased significantly with

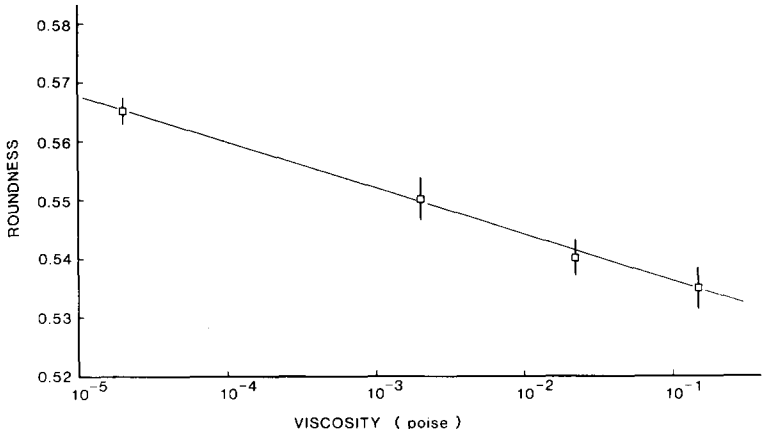

Fig. 5. Effect of fluid viscosity on the particle roudness after 5 h of ball mill grinding.

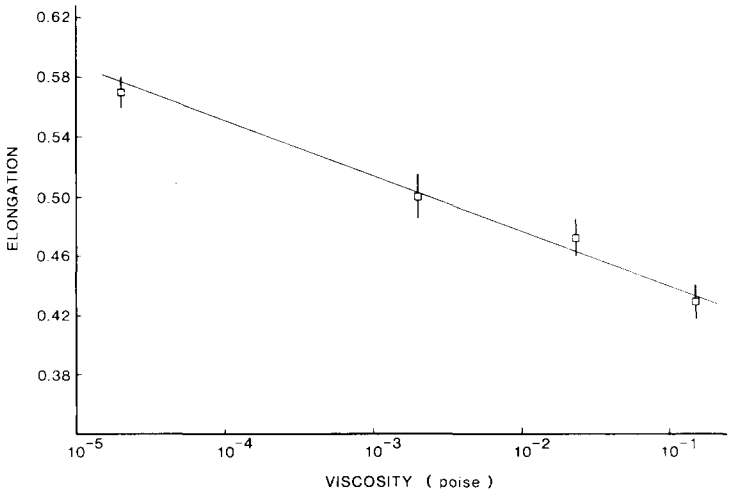

Fig. 6. Effect of fluid viscosity on the particle elongation after 5 h of ball mill grinding.

the increase in the fluid viscosity. The same was noticed for the degree of elongation. The proposed explanation for this behaviour is as follows: when a salicylic acid crystal is crushed in the dry state a large number of cracks will be initiated through the random forces applied on the particle. These cracks produce a large number of faces. Consequently a fragment with a large number of faces is, on statistical grounds more likely to be equidimensional. This can explain the increase in roundness observed in the dry milling of salicylic acid. On the other hand, a particle subjected to ball milling in presence of a liquid medium will become more sensitive to the mode of operation. During wet grinding, apparently, the presence of a liquid as a grinding medium produced a preference in orientation and alignment of the salicylic acid crystals at the single crystal level. The random forces which were predominant in dry milling are modified into more ordered forces. The effective forces used in grinding were reduced because of the resistance of the fluid between the balls. As a result, fracture and loss of corners were substituted by plastic deformation without fragmentation. This gave rise to a more elongated and less rounded particle. The degree of elongation is viscosity-dependent (see Figs. 5, 6).

TABLE 1

Effect of dry and wet milling on the elongation and roundness of salicylic acid crystals reduced to the same particle size (50 μm).

Grinding method	Duration	Elongation	Roundness
Dry milling		0.521243	0.564396
(in air)	30 min	±0.00842	±0.00775
Wet milling		0.443982	0.526574
(in HLP)	5 h	±0.00540	±0.00615

The comparison of the shape parameters of the 50 μm particle size, obtained by size reduction, in dry and wet milling after 30 min and 5 h respectively, indicates clearly that the viscosity of the grinding medium has a direct influence on roundness and elongation (see Table 1). This finding disproves the hypothesis that decrease in particle size generally increases roundedness and elongation.

In view of these results, it appears that homogenization of drugs suspended in liquids of different viscosities for oral or parenteral use could produce dramatic changes in particle morphology. These changes are important in relation to the flow characteristics and the stability of these suspensions. Further research in this area is needed to quantify these effects and to establish the importance of material structure in relation to fracture and morphological changes in the wet-grinding.

Conclusion

Dry ball milling led to the production of particle fragments more round than the original crystals due to the random orientation and collision during the rotation. In presence of liquids, preferred orientation and alignment of salicylic acid crystals produced fragments that are more elongated and less rounded. These changes are due to the plastic deformation and the difficulty of the stressed particles to reach the breaking point. The magnitude of the change in particle shape depends on the viscosity of the grinding media.

Acknowledgements

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