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DETERMINATION OF SPHERICITY OF PELLETS PREPARED BY EXTRUSION/SPHERONIZATION AND THE IMPACT OF SOME PROCESS PARAMETERS.

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ABSTRACT

A method for the quantitative assessment of particle shape has been evaluated and used to study the influence of some process parameters on the sphericity of particles produced by extrusion/spheronization.

The measuring technique is based on the transfer of a microscopic picture of particles to a video screen. From the digitized image, the length/width ratio for a large number of particles is calculated by a computer.

Processing conditions, such as added amount of liquid and spheronization speed/time, have a pronounced impact on particle sphericity.

The results obtained and the convenience of the method demonstrate that the measuring technique is valuable in following changes of particle shape in relation to processing conditions.

INTRODUCTION

One technique for obtaining spherical particles is extrusion and spheronization. The spheronization equipment generally operates by extruding wetted material into cylindrical segments, breaking the segments and then rolling them into solid spheres. The main processing steps are dry blending, wet mixing, extrusion and rolling on a friction plate (1-3).

The plasticity properties of the mass and the values of various processing variables are essential for obtaining a spherical product. A quantitative assessment of the particle shape can give valuable manufacturing information, since the shape can be of importance for subsequent processing steps like coating and packaging.

This paper presents a convenient and reliable method for shape measurement, and characterizes the influence of some processing parameters on the shape.

SHAPE MEASUREMENTS

Method for Shape Measurements

Particle shape can be described by various form factors (see references 4,5). One example is Heywood's shape factor, derived from the ratio between particle surface area and volume (6).

Among various techniques described in recent years for the measurement of differences in shape, are the simulation of use of a tilted plane (two-dimensional) (4), and shape sorting with slotted sieves (three-dimensional) (5).

The method described and evaluated in this work is based on the transfer of a microscopic picture of particles to a video screen. A representative sample of pellets is placed under a magnifying lens, and a picture obtained with a video camera is used for a quantitative analysis of particle dimensions in two planes. A similar technique was recently reported (7).

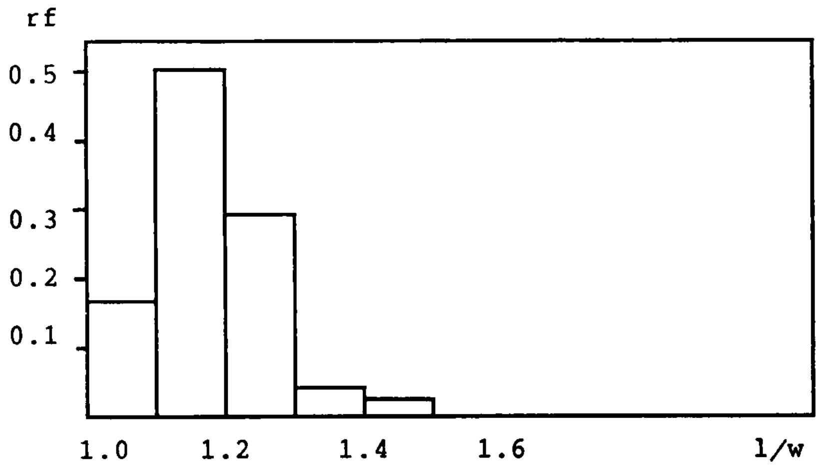
The software, called MicroScale RT, captures and digitizes the picture into 512 x 512 pixels. An 8-bit grey-scale thresholding allows the system to recognize the boundaries of the pellets.

From the captured image the length and width of each particle is measured, using the longest axis as the length (l) and defining width (w) as the measure across the particle on a line perpendicular to the midpoint of the longest axis.

The maximum length and the width are recorded for each particle, and the ratio l/w is calculated. A sample of approximate 400 pellets is divided into classes by the ratio, giving a frequency histogram (see Figure 1).

In an ideal sample, all particles have l/w = 1.0. The more the pellets tend to deviate from ideal spherical shape, the greater the ratio and the more the particle distribution tends toward the right in the histogram.

A conceivable measure of the weighted average for the particle distribution would be the total sum of class midpoint multiplied by the relative frequency for each class. For perfect spheres the proportion of pellets present in the first class will be 1.0. The class midpoint for this class has the value of 1 plus half the class width. With a class width of (e.g.) 0.1, the sphericity value will be [(1+0.05)x1.0]= 1.05. This measure will increase with decreasing sphericity. By inversion of the calculated value and multiplication



rf= realtive frequency (fraction of 1)

FIGURE 1.
Sample of Pellets Sorted by the l/w Ratio into a Histogram.

by 100%, the obtained value will increase with improved shape.

With the intention to have a value of 100% for perfect circularity, the lower class limit (b) has been exchanged for the class midpoint for each class. Thus, the overall sphericity for the sample would be calculated as

$$\frac{1}{\Sigma(b \times rf)} \times 100\% \qquad \text{(Eq. 1).}$$

In trying to characterize different samples with clear differences in particle shape, the derived measure to describe the roundness gave numerical values with small differences. By raising the factor b to the

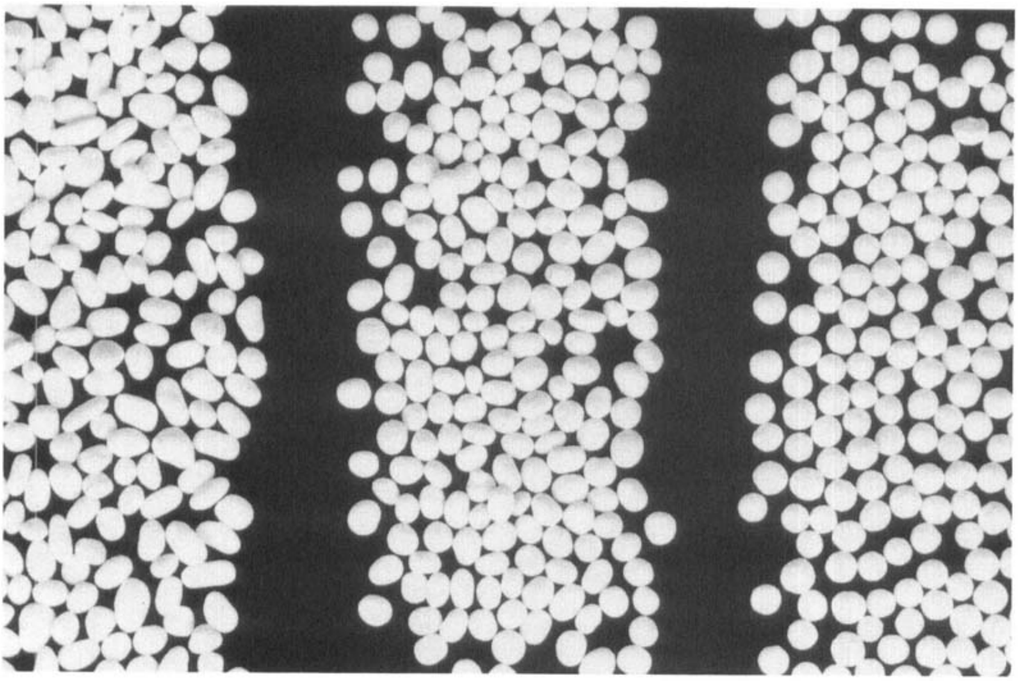


FIGURE 2.

Shapes of Pellets with Different S-Values.

second power, a wider range of calculated shape values is obtained. Thus, the equation used for the sphericity (S) can be written

$$S = \frac{1}{\Sigma(b^2 \times rf)} \times 100\% \qquad \text{(Eq. 2).}$$

The meaning of this value is visualized in Figure 2.

It should be noticed that the resulting S-values will depend on the selected width of the classes.

A wider class width increases the sphericity value for a given particle distribution, whereas the selectivity between different samples decreases.

TABLE 1.

Reproducibility of Method. Sphericity Values, S %.
(Using class width= 0.1.)

	Batch				
	A	B	C	D	E
Occasion 1	74.7	74.5	76.0	80.1	68.2
Occasion 2	72.3	77.1	77.9	83.3	70.7
ΔS	2.4	-2.6	-1.9	-3.2	-2.5

Standard error calculated by

$$SE = \sqrt{\sum_{i=1}^{k} (x_{i1} - x_{i2})^2 / 2k}$$, as given by G. Blom (8).

$x_{i1} - x_{i2} = \Delta S$ and k= number of samples

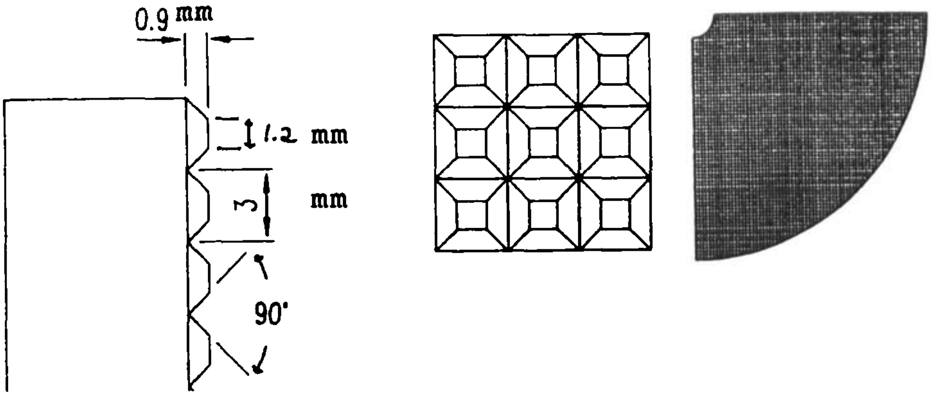
$$SE = \sqrt{\frac{5.76}{10} + \frac{6.76}{10} + \frac{3.61}{10} + \frac{10.24}{10} + \frac{6.25}{10}} = \sqrt{3.262} = 1.81$$

Precision of the Method of Measurement

The repeatability was tested by analyzing the same sample three times. The sphericity values for the pellets were 76.9%, 76.4% and 77.6%, giving a coefficient of variation of 0.8%.

Five batches of pellets prepared under somewhat different conditions were each analyzed on two separate occasions to give information about the reproducibility of the method. The result is shown in Table 1 below. As the measurements were made on two separate samples from each batch, a contribution to the standard error caused by possible inhomogeneity of each batch will be included in the figures. However, this contribution is small, as shown in Table 1.

FIGURE 3.



Indentation Pattern of the Friction Plates.

EXPERIMENTAL

Preparation of pellets

Throughout the study, the same formulation was used, mainly consisting of a sugar and a model drug substance. The ingredients were dry mixed for 1.5 min in an intensive mixer (Eirich R 02). Water, the granulation liquid, was added under constant conditions by a peristaltic pump (Watson-Marlow model 601).

The wet mass was pressed through a twin-screw radial extruder (Fuji Paudal EXD-60) fitted with screen plates, apertures 1.2 mm in diameter.

The extruded cylindrical segments were transferred in weighed portions to spheronizing plates (built at

Hässle AB) of varying diameters. The pattern of the plates was the same and is shown by Figure 3.

After spheronization the pellets were dried for 20 min in a fluidized bed dryer at 60°C, and the sieved fraction 1.0-1.6 mm was collected (Vibro, Retsch KG). A representative sample was obtained by dividing the resulting pellets with a spinning riffler.

Studied Process Variables

For pellets manufactured with the described technique, the assessment of two-dimensional data will give adequate information for optimization of particle shape in three dimensions.

Changes in processing conditions were made in the wetting step, the extrusion as well as in the spheronization, after which particle sphericity was determined.

In the experiments the amount of granulation liquid was alternated between 17% and 23%, calculated on the weight of dry ingredients. These limits were set considering that the moisture range should be as large as possible without getting a mass that was impossible to process.

In all trials, the water was added during 1.5 minutes followed by wet mixing, normally during 3 minutes.

The wetted material was extruded and the first material out of the extruder was discarded in all experiments.

Spheronization was normally made in portions of 400 g of extrudate. The material was treated on a friction plate with a diameter of 42 cm for 3 min at 350 rpm. This spheronization procedure was used as a standard operation.

TABLE 2.
Dwelling Time Experiments. Plate Load = 400g.

Added water, % :	17		23	
Extrusion speed, rpm:	44		36	
Spheronization;				
speed, rpm :	350		350	
plate diameter,cm :	42		42	
dwelling time, min :	3	7	3	7

Changes in particle shape in relation to high and low water content were evaluated according to this procedure.

The effect of varying wet mixing time was studied in experiments performed with the same procedure. Three batches moistened with 23% of water were wet mixed for 2, 4 and 9 minutes, respectively. The influence of the water content and mixing time is given in Table 5.

The impact of extrusion speed was tested by changing the feed rate between 706 g/min and 1492 g/min. This corresponds to 15 rpm and 64 rpm, the limitations of the equipment. The formulation was examined at both moisture levels, and standard spheronization operations were used. Effect on sphericity is shown in Table 6.

The influence of dwelling time and added amount of liquid was studied according to Table 2.

Table 3 shows the processing levels used for the study of the influence of plate speed and diameter on the sphericity.

An increase in the amount of extrudate placed into the spheronizer was made in separate trials. Charges of

TABLE 3.

Variation of Spheronization Speed. Plate Load= 400g.

Added water, % :			23		
Extrusion speed, rpm:			25		
Spheronization;					
dwelling time, min :			3		
speed, rpm :	317	350	450	456	668
plate diameter, cm :	42	42	42	32	22

TABLE 4.

Experiments With Different Charges. Plate Diameter= 42 cm.

Added water, % :	17		23	
Extrusion speed, rpm:	36		44	
Spheronization;				
dwelling time, min :	5		5	
speed, rpm :	350		350	
charge, g :	400	1000	400	1000

400g and 1000g were tested, each at two extrudate moisture levels, 17% and 23% (see Table 4).

RESULTS AND DISCUSSION

Parameters investigated in the wet mixing step were amount of added liquid and wet mixing time. The results are listed in Table 5.

TABLE 5.

Influence of Added Water and Mixing Time on Pellet Sphericity. Extrusion Rate 15 rpm.

Amount of liquid %	23	17	23	23	23
Wet mixing time,min	2	3	3	4	9
Spheronization		standard		operation	
Sphericity (S)	79.9%	67.7%	80.6%	77.9%	75.5%

The amount of added liquid has a significant effect on the sphericity of the pellets. Increasing addition of water improves the moulding properties of the mass so that the extrudates are more easily and effectively rounded to spherical pellets.

When wet mixing time is considered, it is obvious that an extended treatment affects the powder mixture and deteriorates the properties of the mass. This can be observed as an influence on the measured sphericity, expressed as a diminishing shape value with prolonged wet mixing time.

In the extrusion step, several processing parameters can be changed and the equipment modified, which may have an effect on the roundness of the final product. The screen hole diameter, the hole area of the total screen area, the ratio between hole diameter and screen thickness, screen thickness, frontal or radial extrusion, extrusion rate, extrusion pressure and screw configuration can be mentioned.

In this work, only the effect of high and low extrusion rate have been tested. This is done at the two extreme levels of moisture content. Table 6 shows the results obtained.

TABLE 6.
Influence of Extrusion Rate on Pellet Sphericity.

Extrusion rate, rpm :	15	64	15	44
Amount of liquid, % :	23	23	17	17
Wet mixing time, min:	4	4	3	3
Spheronization :	stand.	stand.	stand.	stand.
Sphericity (S) :	80.1%	79.9%	67.7%	67.2%

It is obvious that the formulation is insensitive to extrusion output rate. The velocity did not appear to be critical, and possible pressure changes have minor effects on the properties of the mass. The same shape values were obtained regardless of extruder speed within the specified water levels. However, the related improvement of sphericity with increasing liquid addition is clearly illustrated, which is in agreement with previously obtained results (see Table 5).

The effect of rolling the extrudate on the friction plate during periods of different length was tested with two masses with different water contents, giving different rheological properties. According to previous experiments, the mixture with 23% water can be regarded as optimal for the formulation used, considering its consistency, processability and ability to form well-shaped spherules. Additional liquid gives a mass that is impossible to process in the equipment used.

The results in Table 7 show that the impact of spheronization time is more pronounced for the dry formulation. After 3 minutes' dwelling time, the pellets derived from the less moistened mass have the most irregular shape. The S-value was calculated to be 67.2%.

TABLE 7.
Influence of Dwelling Time on Pellet Sphericity.

Amount of liquid, %:	17		23	
Dwelling time, min :	3	7	3	7
Sphericity (S) :	67.2%	79.7%	76.1%	79.7%

At the higher moisture level, the same short processing time was enough to give a much better shape, S= 76.1%.

Longer dwelling time results in pellets with higher particle regularity, and the shape factors for these two experiments were found to be identical.

In the literature (Woodruff et al. 2), it has been postulated that longer processing time leads to impaired sphericity due to surface drying, with subsequent erosion of the particle surface. The opposite effect presented above in this paper has also been described by other investigators (4). These contradicting results can possibly be related to the formulations investigated.

The importance of spheronization speed and friction plate diameter for particle shape were illustrated with five experiments summarized in Table 8.

The indentation of the plates shown in Figure 3 was the same for the three plates studied.

The results obtained using a plate diameter of 42 cm show that there is a direct relationship between plate speed and resulting sphericity. An increased speed gives, as expected, more regular and rounded

TABLE 8.

Influence of Plate Speed on Pellet Sphericity.

Plate diameter, cm	42	42	42	32	22
Plate speed, rpm	317	350	450	456	668
Peripheral vel. m/s	7.0	7.7	9.9	7.6	7.7
Sphericity (S)	77.6%	80.7%	87.4%	81.8%	82.4%

particles. Centrifugal and frictional forces on the serrated plate become higher with increased peripheral velocity, and together with interparticulate friction of the moving pellets, more effective shape forming conditions are created.

When the pelletizations with different plate diameters are compared, it seems that the same peripheral velocity leads to a similar appearance for the pellets produced, regardless of plate size. The observed S-values lie between 80.7 - 82.4 % at a speed of 7.7 m/sec. These results were obtained using the same amount of extrudate on the different plates.

As can be seen in Figure 4, a good correlation between plate peripheral velocity and pellet sphericity values is obtained, although plates of different diameters are used.

The treatment on the friction plate contributes significantly to the pellet shape. The assumption that different amounts of extrudate on the plate can cause changes in the shape obtained is plausible, as long as other conditions do not overcome the effect of increase or decrease in load size. Results from tests with different loads on plate of diameter 42 cm are given in Table 9.

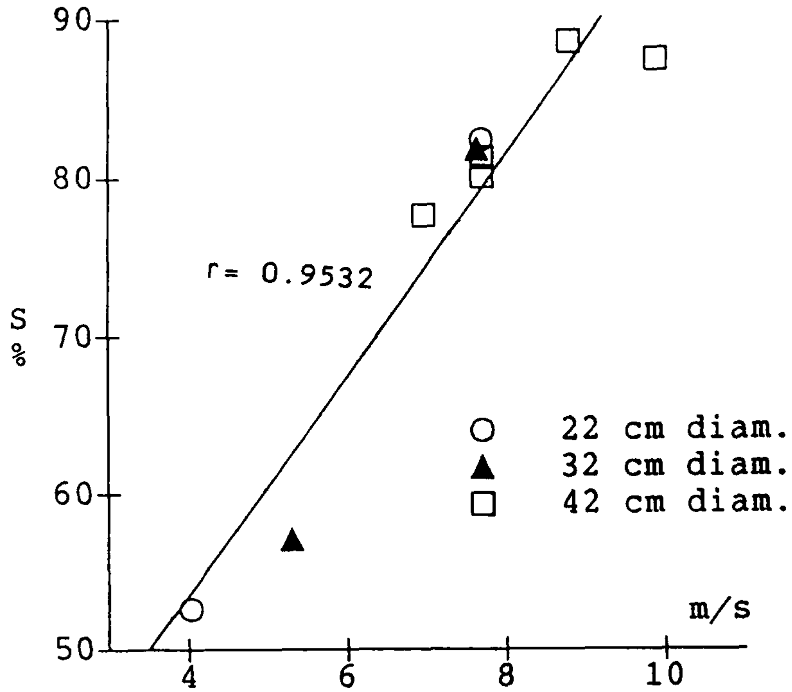


FIGURE 4.

Relationship Between Plate Periphery Velocity and Sphericity.

TABLE 9.

Influence of Amount of Extrudate on the Obtained Pellet Sphericity. Dwelling Time on Plate = 5 min.

Charge, g :	400	1000	400	1000
Added water, %:	17	17	23	23
Sphericity (S):	74.5%	73.1%	80.1%	74.7%

TABLE 10.

Variation Between Batches Produced With the Same Parameter Settings. (Wet mixing time 3 min., 23% moisture content, extrusion rate 15 rpm and spheronization conditions at standard levels on the 42 cm plate.)

Experiment nr :	1	2	3	4	5	6
Spericity (S) :	80.1%	81.3%	80.1%	79.9%	80.6%	79.9%

Average value= 80.3%

Coefficient of variation =0.68%

The spheronization of pellets depends on the energy transferred from the plate as friction and motion. The amount of energy transferred to each particle within a specified time decreases with increasing load.

This reduction has a negative influence on the shape-forming process. This can be seen for the damp mass with strong inherent cohesive forces.

The dry mass of the formulation studied seems to be less sensitive regarding shape, when different loads are spheronized, since no impairment of the shape was observed when the load was increased. This may be explained by a compensatory increase in interparticulate friction, which affects the surface erosion of these more fragile extrudates.

When processing conditions are established for a given formulation, many product characteristics must be considered. In all manufacturing it is desirable to achieve a high homogeneity for all product properties.

For a pellet product, it is important that the shape is reproducible from batch to batch. This has been tested on six batches, and the results are presented in Table 10.

CONCLUSIONS

Sphericity is related to the processing conditions used in the manufacturing of pellets. For the formulation studied, the amount of added liquid has a pronounced effect on the shape of the pellets. An extended wet mixing time results in shape impairment, while the speed with which the material is forced through the extruder does not seem to influence the roundness.

A good positive correlation between the plate peripheral velocity and the sphericity is observed. Dwelling time on the plate has an appreciable effect on the shape. The amount of extrudate on the plate can have an impact on particle sphericity.

The method used in this work to characterize the roundness is a preferable alternative to other described two-dimensional techniques, i.e. less time consuming and/or more representative. Precision, measured as repeatability and reproducibility, was high and the method itself provides a feasible way to evaluate the impact of processing parameters on the sphericity of pellets.

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