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The evaluation of a three-dimensional shape factor for the quantitative assessment of the sphericity and surface roughness of pellets

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Abstract

A three-dimensional shape factor for the characterisation of the quality of pellets has been developed based on image analysis. The shape factor includes the separate assessment of deviations from the spherical shape and the presence of surface roughness. These two components can be viewed and compared with the overall three-dimensional shape factor e_{c3}, which connects the two features. The shape factor e_{c3} has been applied to three sets of pellets prepared by extrusion/spheronisation, and a comparison with the Heywood shape factors has been undertaken. The results demonstrate the ability of e_{c3} to differentiate various batches of pellets based on a defined mathematics, whereas the Heywood shape factors require certain assumptions about the particle outline to be applicable.

Keywords: Image analysis; Particle shape; Pellet; Sphericity; Surface roughness

1. Introduction

The exact definition of particle shape is still a problem to be solved in powder technology. Precise mathematical definitions are possible for fixed particle shapes such as a rectangular parallelepiped, pyramids or cylinders (see Documenta Geigy, 1962), but common particles seldom follow precisely such shapes. Hence, the actual shape of a particle can only be judged in comparison to an ideal shape, and an overall shape factor based on a fixed mathematical definition appears impossible.

The spherical shape is of special interest in extrusion/spheronisation and other pelletisation techniques. Therefore, several studies have investigated the possibility to describe the deviations from being a sphere using the two-dimensional particle outline obtained from photography, microscopy or image analysis. Up to 20 different shape factors (Yliruusi et al., 1992) have been devised and compared without presentation of a satisfactory solution. Most popular are the aspect ratio (Schneiderhöhn, 1954), which is the ratio between the longest distance of a particle and its perpendicular dimension, and the elongation ratio (Tsubaki and Jimbo, 1979), which is the ratio between the smallest Feret diameter and its per-

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pendicular Feret diameter. In principle, these two descriptors provide different numbers for irregular shaped particles. Some authors (Lindner and Kleinebudde, 1993) were only concerned with the assessment of shape of spherical particles, and hence came to the conclusion that aspect and elongation ratio are mathematically equivalent. However, the process of extrusion/spheronisation can lead to irregular as well as spherical particles, and hence a clear definition is essential if comparisons are to be made. Beddow (1983) compared the information content of several shape factors and concluded that aspect and elongation ratio do not reflect truly the shape of a particle. Hence, they should be avoided. Another popular shape factor for spherical particles is called circularity, first defined by Cox (1927), and later redefined by Hausner (1966) as the reciprocal of the Cox value. Again, the circularity is unable to differentiate between symmetrical figures such as squares or circles providing in both cases the numerical value of 1.

Problems in powder behaviour such as uneven powder flow cannot be predicted from a two-dimensional particle shape factor, because the volumetric structure of the particles in a powder bed causes such problems. Hence, three-dimensional shape factors are desirable. In this respect Heywood (1954, 1963) developed three-dimensional shape factors, which are based on empirical equations. Air permeametry is another possible method to assess the three-dimensional shape of particles (Robertson and Emödi, 1943; Eriksson et al., 1993). However, air permeability measurements only provide an average shape factor of the particles in the powder bed and give no information about the distribution of shape.

The shape and surface texture of a spherical particle are two independent parameters (Hawkins, 1993), which need separate assessment by image analysis. Surface texture can be expressed as the ratio between the true perimeter of a two-dimensional particle outline, which includes all surface roughnesses, and a convex hull surrounding the particle outline (Barrett, 1980). A different method was proposed by Podczeck and Newton (1994), which is based on the comparison of the perimeter with an average radius equivalent circle. For the case of a two-dimensional projection of the particle outline, the authors were able to combine shape and texture to a single shape factor for the assessment of sphericity, and the aim of this work is to extend their two-dimensional into a three-dimensional description of the particle shape and surface texture. The efficiency of the three-dimensional shape factor will be compared with the classical three-dimensional shape factors described by Heywood (1963).

2. Theory

To assess the three-dimensional particle outline using image analysis, a spheroid needs to be inspected from two different points, which are placed 90° to each other, and which deliver two two-dimensional images of the particle shape outline (see Fig. 1). The usual problem of manufacturing spheres by extrusion/spheronisation is that the shape of the pellets can deviate from that of a sphere, usually resulting in a product which is in the shape of an ellipsoid (Newton, 1994). Hence, an appropriate shape factor should be based on

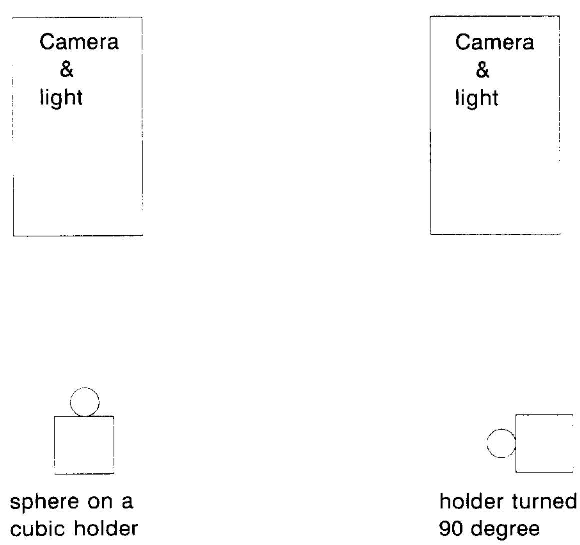

Fig. 1. Three-dimensional image analysis of a spherical particle.

the linear eccentricity (e) of an ellipsoid, which is defined as:

$$e = \sqrt{(a^2 - b^2) + (a^2 - c^2)} \quad (1)$$

where a = distance between the centre of gravity and the surface along the main axis ($2a$ = length of the ellipsoid), and b and c = distances between the centre of gravity and the surface along the two secondary axes perpendicular to the main axis ($2b$ = breadth of the ellipsoid, $2c$ = thickness of the ellipsoid). In the case of a perfect sphere ($a = b = c$) the value for e is zero.

The numerical value of the linear eccentricity of an ellipsoid depends on the numerical values of the three axes and therefore a normalization is required, i.e., the values of the two short axes are divided by the value of the main axis. Since length, breadth and thickness of the ellipsoid are related to their equivalent axis by a factor of 2, which disappears due to the normalization, the normalized equation for the linear eccentricity can be written as:

$$e_n = \sqrt{\left(1 - \left(\frac{b}{l}\right)^2\right) + \left(1 - \left(\frac{t}{l}\right)^2\right)} \quad l \geq b \geq t \quad (2.1)$$

where l = length, b = breadth and t = thickness of the ellipsoid. Eq. 2.1 can be simplified to:

$$e_n = \sqrt{2 - \left(\frac{b}{l}\right)^2 - \left(\frac{t}{l}\right)^2} \quad l \geq b \geq t \quad (2.2)$$

For a two-dimensional image, an estimate of the surface texture ('roughness') can be found, if the distances between the centre of gravity of the particle outline and the perimeter are determined measuring a series of distances which differ in an angle α from each other (Podczeck and Newton, 1994). The mean value of these 'radii' is equivalent to the radius of a perfect sphere, but surface irregularities cause a shortening of the value, and the perimeter of the pellets will be underestimated. Therefore, in the two-dimensional case, the surface roughness was determined from (Podczeck and Newton, 1994):

$$s_r = \frac{2 \cdot \pi \cdot r_e}{P_m} \quad (3)$$

where s_r = surface roughness, r_e = arithmetic mean of the distances between the centre of gravity and the perimeter for a given α between the measurements and P_m = perimeter of the particle outline. A perfect sphere has a surface roughness value of 1, and surface roughness leads to values smaller than 1.

For an elliptical two-dimensional particle outline, this surface roughness value needs a correction, because the elipticity itself leads to an apparent surface roughness r_s, which is about 0.8 for a normalized ellipse with a breadth of 0.2 (length = 1). However, the effect of elipticity on the surface roughness can be described as a linear function ($r = -0.998$, root mean square of the residual analysis: rms = 2.0%), which is valid both for normalized and unstandardized ellipses:

$$f = 1.008 - 0.231\left(1 - \frac{b}{l}\right) \quad (4)$$

where the thickness t can replace the breadth b, and where f is a factor, which can be used to correct the surface roughness value as follows:

$$s_r = \frac{2 \cdot \pi \cdot r_e}{P_m \cdot f} \quad (5)$$

For the three-dimensional shape factor the surface roughness can roughly be estimated applying Eq. 5 to the two two-dimensional particle outlines measured (see Fig. 1). The three-dimensional shape factor combines surface roughness and eccentricity as follows:

$$e_{c3} = \frac{\left(\frac{2 \cdot \pi \cdot r_e}{P_m \cdot f}\right)_1 + \left(\frac{2 \cdot \pi \cdot r_e}{P_m \cdot f}\right)_2}{2} - \sqrt{2 - \left(\frac{b}{l}\right)^2 - \left(\frac{t}{l}\right)^2} \quad (6)$$

where the subscripts 1 and 2 indicate the two two-dimensional particle outlines measured.

It is advisable to list the shape factor e_{c3} and the surface roughness r_s $[r_s = ((2\pi r_e/(P_m f))_1 + (2\pi r_e/(P_m f))_2)/2]$ together to allow a separate judgement of particle shape and particle surface texture.

A perfect sphere has a three-dimensional shape factor of $e_{c3} = 1.0$, whereas rough spheres or non-spherical particles have a shape factor smaller than 1.0.

3. Materials and methods

Three batches of pellets were prepared by extrusion/spheronisation, varying the proposed particle shape by changes in the formula used. Furthermore, the pellets used for this study were preselected from the original batches to give three samples of defined shape –visually spherical, oval and irregular particles. The size (length) of the pellets was constant in each sample.

The pellets were mounted on wooden, rectangular sticks ($50 \times 2 \times 2$ mm), which had been coated with black paint. Care was taken to fix the pellets in the most stable position ('critical stability' position according to Heywood (1963)). It is necessary to provide supports which are wider than the pellets, because the holders need to be turned by 90° (see Fig. 1), and pellets larger than the support would therefore touch the surface making a complete turn impossible.

The measurements were carried out using an Image Analyzer (Seescan solitaire 512, Seescan, Cambridge, UK), completed with a black/white camera (CCD-4 miniature video camera module, Rengo Co. Ltd, Toyohashi, Japan) and a zoom lens (18-108/2.5, Olympus Co., Hamburg, Germany). A cold light source (Olympus Co., Hamburg, Germany) was used in top light position to illuminate the pellets against a black surface.

Pellets were placed in the field of view in each of the two positions at right angles to each other (see Fig. 1) to provide two sets of raw data of their shape. 15 pellets of each proposed particle shape were analyzed, and the output file contained length, breadth, thickness, the perimeters of the first and second assessment and the estimated perimeters (based on 360 radii per two-dimensional particle outline) according to Eq. 5 (see Section 2). After file transport the shape factor e_{c3}, its two components s_r and e_n, and the Heywood surface and volume shape factors, f and k, respectively, were calculated on a PC.

4. Results and discussion

Podczeck and Newton (1994) encountered some problems in finding the ideal sphericity for ball bearings, which are supposed to be perfectly round. One reason for this was the illumination technique they had used, which was the classical way of transmission of light. Ball bearings have a polished surface which reflects the light diffusely and therefore the outer surface becomes less sharp and overshadowed (Foley and Van Dam, 1982). Furthermore, image analysis tries to compose any shape from a linear raster of pixels, which transforms any curved line into a stepped line. Additionally, dark objects against a bright background have less sharp contours because of a finite change of the signal of a monochrome camera (Lindner and Kleinebudde, 1993). Hence, a different illumination technique was used (see Section 3). The ball bearings were coated with white paint. Now there were white objects against a black background. There was still diffuse light reflection, but the resulting shadow was black on a black background and therefore invisible. The e_{c3} value for the ball bearings using the previous illumination technique was about 0.4, whereas the illumination technique used in this study provided a reference value of 0.722 ± 0.016. The difference from the ideal e_{c3} value (1.0) of 27.8% can be attributed to the problem of linear pixel rasters discussed above and especially to the coating of the ball bearings.

Table 1 compares the shape factor e_{c3}, its components s_r (roughness of the surface) and e_n (linear eccentricity) and the Heywood shape fac-

Table 1
Shape descriptors for three samples of pellets

	Spherical	Oval	Irregular
e_{c3}	0.366 ± 0.106	0.262 ± 0.093	-0.130 ± 0.120
s_r	0.908 ± 0.023	0.919 ± 0.024	0.898 ± 0.044
e_n	0.543 ± 0.104	0.657 ± 0.096	1.001 ± 0.102
f	2.998 ± 0.039	2.905 ± 0.113	2.517 ± 0.204
k	0.514 ± 0.018	0.498 ± 0.047	0.437 ± 0.077

e_{c3}, three-dimensional shape factor; s_r, surface roughness component of e_{c3}; e_n, linear eccentricity component of e_{c3}; f, Heywood surface shape factor; k, Heywood volume shape factor.

tors f (surface factor) and k (volume factor). The shape factor e_{c3} for the proposed spheres (0.366 ± 0.106) is only half of the value obtained for coated ball bearings (0.722). Indeed, the image of these pellets suggested a polyhedral shape instead of a perfect sphere. The flattening of the surface could be the result of an insufficient duration of processing of the extrudate in the spheroniser and leads to a drop of the numerical value of the shape factor. The Heywood surface shape factor f for a perfect sphere is equal to the constant π (≈ 3.142). The difference between this value and that obtained for the proposed spheres (2.998 ± 0.039) is statistically significant ($t = 14.26$; $t_{14;\ P=0.05} = 2.14$) again suggesting that these pellets are not really spherical.

The very low value of e_{c3} for irregular pellets is not very much due to surface roughness as the definition of irregularity might suggest. Again inspection of the images has shown that the uneven shape is a result of flattening, but that the single edges of the pellets are quite smooth. Spheronisation breaks the cylindrical extrudate into small lengths which then are rounded (Newton, 1994). The irregularity is a result of inefficient breakage, probably due to moisture and stickiness leading to a large value for e_n. The densification of the mass during extrusion is responsible for the surface roughness, and the formula chosen behaves well in this respect. Therefore, surface roughness does not affect the shape factor very much as can be seen from the value of s_r. It might be worth

Table 2
Statistical analysis of shape factors obtained from three batches of pellets produced by extrusion/spheronisation

e_{c3} (analysis of variance)				
$F = 79.64$	pair comparison	$F_{tab} = 4.20$	($f_1 = 1$; $f_2 = 28$)	
$F_{tab} = 3.23$	sphere-oval	$F = 7.03$		
($f_1 = 2$; $f_2 = 42$)	sphere-irregular	$F = 144.28$		
	oval-irregular	$F = 87.62$		
e_n (analysis of variance)				
$F = 84.70$	pair comparison	$F_{tab} = 4.20$	($f_1 = 1$; $f_2 = 28$)	
$F_{tab} = 3.23$	sphere-oval	$F = 9.73$		
($f_1 = 2$; $f_2 = 42$)	sphere-irregular	$F = 156.33$		
	oval-irregular	$F = 88.05$		
s_r (variance inhomogeneity)				
Bartlett test	pair comparison (Welch test)			
$\chi^2 = 109.51$	sphere-oval	$t' = 1.28$	$v = 27$	$t_{tab} = 2.05$
$\chi^2_{tab} = 5.99$	sphere-irregular	$t' = 0.78$	$v = 21$	$t_{tab} = 2.08$
	oval-irregular	$t' = 1.62$	$v = 21$	$t_{tab} = 2.08$
f (variance inhomogeneity)				
Bartlett test	pair comparison (Welch test)			
$\chi^2 = 29.02$	sphere-oval	$t' = 3.01$	$v = 17$	$t_{tab} = 2.11$
$\chi^2_{tab} = 5.99$	sphere-irregular	$t' = 8.97$	$v = 15$	$t_{tab} = 2.13$
	oval-irregular	$t' = 6.44$	$v = 21$	$t_{tab} = 2.08$
k (variance inhomogeneity)				
Bartlett test	pair comparison (Welch test)			
$\chi^2 = 22.99$	sphere-oval	$t' = 1.23$	$v = 18$	$t_{tab} = 2.10$
$\chi^2_{tab} = 5.99$	sphere-irregular	$t' = 3.77$	$v = 15$	$t_{tab} = 2.13$
	oval-irregular	$t' = 2.62$	$v = 23$	$t_{tab} = 2.07$

e_{c3}, three-dimensional shape factor; s_r, surface roughness component of e_{c3}; e_n, linear eccentricity component of e_{c3}; f, Heywood surface shape factor; k, Heywood volume shape factor; F, test value analysis of variance; f_1, f_2, 1st and 2nd degree of freedom; F_{tab}, tabulated F at 5% level; χ^2, test value Bartlett test; χ^2_{tab}, tabulated χ^2 for 2 degrees of freedom at 5% level; t', test value Welch test; v, degree of freedom Welch test; t_{tab}, tabulated t value at 5% level.

pointing out that Hawkins (1993) has investigated the reproducibility of polygonal shaped particle outlines using polar radii. Major surface roughnesses could be detected already with an angle of 10° between the single lines. In this study the angle between the radii was 1° (see Section 3) and therefore it can be assumed that a true surface roughness should have been detected.

One point of interest is usually the differentiation between batches of spherical pellets in terms of their true shape. Hence, the shape factors listed in Table 1 were used for a statistical analysis. First, the values of e_{c3}, and e_n were used in an analysis of variance. All three samples are statistically different in shape and linear eccentricity (see Table 2). There is severe variance inhomogeneity between the values for r_s, f and k (Bartlett test, see Table 2) and therefore analysis of variance is not applicable. However, in such cases a Welch test (see Snedecor and Cochran, 1980) can be used to compare the mean values. This test showed that the surface roughness r_s is not different for the three pellet samples tested. The Heywood surface factor f differentiated between all three pellet samples (see Table 2). Both e_{c3} and f indicate that the largest difference in shape is between the spherical and the irregular pellet batch, as expected. The difference between the proposed spheres and the oval pellets is also clearly reflected in the shape factors e_{c3} and f. Using only the values of length and breadth, for the oval and spherical sample an aspect ratio could be calculated which was 1.087 ± 0.055 and 1.062 ± 0.043, respectively. The aspect ratio would therefore fail to distinguish between the two batches of pellets ($t = 1.39$, $t_{28,P=0.05} = 2.05$). The Heywood volume factor k was also not able to detect a difference between the spherical and the oval pellets (see Table 2). The difference between the irregular pellets and the two other samples is, however, reflected in the significance of the Welch tests for k. The Heywood shape factors are based on two empirical constants, which are variable for different proposed shapes. That means, however, that a change in shape consequently requires a change in the value of the constants, which is practically impossible. Therefore, the factors might fail to detect small shape variations.

The Heywood shape factors and the e_{c3} shape factor are directly proportional. This validates the results obtained for e_{c3}, and it can be concluded that the three-dimensional shape factor e_{c3} is a possible descriptor for deviations in shape for pellets, which are supposed to be spherical.

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