

A general model for hydrocyclone partition curves

M. Frachon, J.J. Cilliers*

Department of Chemical Engineering, UMIST, P.O. Box 88, Manchester M60 1QD, UK

Received 29 September 1998; received in revised form 3 February 1999; accepted 9 February 1999

Abstract

The partition curve is used to quantify the classification in hydrocyclones. It is generally monotonic and asymptotes to a value called the bypass. A ‘fish-hook’ curve occurs sometimes when partition values lower than the bypass are observed. A number of attempts have been made to develop fish-hook partition curve models. These are reviewed and shown to reduce to a general form. The general equation consists of two competing effects: that of classification, described by a conventional corrected partition curve, and that of dispersion, described by an inverse corrected partition curve that is applied to only the bypass fraction. A turbulence model for two-phase systems that quantifies the relative effects of dispersion and classification is described and shown to be applicable to this system. This allows some physical interpretation of the effect of variables on the observed performance. A series of small diameter hydrocyclone experiments illustrate the use of the model and are used to evaluate the bypass. It is often assumed that the bypass can be estimated accurately from the fraction of water in the feed that reports to the coarse product stream. It was, however, found that the recovery of water to the underflow was significantly lower than either the lowest point of the partition curve or the value of the bypass. Further work is required to conclusively determine the variation in the bypass with operating conditions. © 1999 Published by Elsevier Science S.A. All rights reserved.

Keywords: Partition curve; Hydrocyclones; Bypass

1. Introduction

The partition curve is used to quantify and compare the performance of classification in hydrocyclones. In general, the partition curve decreases monotonically with size and asymptotes to a given value called the bypass. A ‘fish-hook’ is sometimes observed in the partition curve when partition values lower than the bypass are observed for a range of particle sizes [1]. This phenomenon has received greater attention since the use of laser-based particle size analyzers allowing the distributions in finer particle sizes to be accurately measured. The mechanism causing the fish-hook has, however, not been explained satisfactorily.

A number of attempts have been made to model partition curves that exhibit a fish-hook. These will be reviewed. It will be shown that they reduce to the same mathematical form and that a general model can be developed. It is often assumed that the bypass can be estimated accurately from the fraction of water in the feed that reports to the coarse product stream. A series of experimental results will be shown to illustrate the use of the model and to evaluate the bypass.

2. Previous work

2.1. The partition curve

The partition curve (also called performance curve, efficiency curve or Tromp curve) shows the fraction of a material of a specific size d in the feed to a classifier that reports to the coarse product stream. In the case of hydrocyclones, this is the underflow. The graph is usually plotted on a log-linear scale to emphasize the fine particle sizes.

For any particle size d the partition number $P(d)$ is calculated from

$$P(d) = \frac{Uu(d)}{Ff(d)} \quad (1)$$

where U and F are the mass flow rates of solids (in the same units) and $u(d)$ and $f(d)$ are the weight fractions of particle size d in the underflow and feed streams, respectively. For hydrocyclones, the partition number for small particle sizes generally is not zero, but asymptotes to a particular value. This value is commonly referred to as the bypass, B_p , which corresponds to the fraction of particles in the feed that bypasses the classification.

If the experimentally measured partition curve is monotonic, it can be corrected to asymptote to zero at small

*Corresponding author. Tel.: +44-161-200 4353; fax: +44-200 4399.

particle sizes [2] by scaling the partition number. The relationship between the experimental partition curve, $P(d)$, the corrected partition curve, $C(d)$, and the bypass, B_p , is given by

$$C(d) = \frac{P(d) - B_p}{1 - B_p} \quad (2)$$

At least two parameters are required to describe a corrected partition curve mathematically. The corrected cut size (d_{50c}) is that particle size at which 50% of the particles of that size in the feed will report to the coarse product stream and the other 50% to the fine product stream. The second parameter is a measure of the shape of the curve, indicating the sharpness of separation.

Corrected partition curves are generally modelled using sigmoidal functional forms. A commonly used equation is the Rosin–Rammler [3,4]

$$C(d) = 1 - \exp\left(-0.693\left(\frac{d}{d_{50c}}\right)^m\right) \quad (3)$$

where m is a measure of the shape of the curve. Other equation forms, such as the exponential sum [5], have also been used.

The general approach to the modelling of hydrocyclone results is to find the parameters of the corrected partition curve and the bypass so that the measured partition curve is closely matched. The values of these parameters under various operating conditions can then be correlated and used to predict the separation performance.

2.2. Modelling the bypass B_p

It is generally assumed [3,6–8] that the bypass is independent of particle size and equals the water recovery from the feed to the underflow, R_f , i.e., $B_p = R_f$.

This assumes that the fraction of the water in the feed recovered in the underflow stream carries with it an equivalent fraction of the feed solids. Austin and Klimpel [9] argue that there is no fundamental reason why, in general, this should be so, and show data where the bypass is clearly not equal to the water recovery.

Svarovsky [10] and Braun and Bohnet [11] assume that the bypass equals the fraction of the feed slurry reporting to the underflow. This assumption is not commonly used, but is a close approximation to the water recovery at low feed solids concentrations and is more readily measured.

2.3. Modelling the fish-hook

When the partition curve displays a fish-hook, the simple ‘correction’ approach outlined above cannot be applied, and a more complex model is required. Eq. (2) can be rearranged to yield

$$P(d) = C(d) + B_p[1 - C(d)] \quad (4)$$

This indicates that the observed partition curve, $P(d)$, can be considered as consisting of two components, $C(d)$ and an inverse partition curve $1 - C(d)$ multiplied by B_p . For a monotonically decreasing partition curve, the two $C(d)$ terms are the same. However, this need not be so, and by changing the form of the second term, a fish-hook shape can be reproduced.

Finch [1] first used this concept and considered the second term, $a(d)$, as a size dependent entrainment function.

$$P(d) = C(d) + a(d) \quad (5)$$

where

$$a(d) = R_f \left(1 - \frac{d}{d_0}\right)$$

The value of d_0 represents the largest particle size affected by the fish-hook. It can be seen that the assumption of the bypass equalling the water recovery to underflow is made, and that dispersion increases as the particle size decreases.

Del Villar and Finch [12] modified this function, arguing that the contribution of the entrainment term is probabilistic and not truly additive. Eq. (5) then becomes

$$P(d) = a(d) + [1 - a(d)]C(d) \quad (6)$$

which can be rearranged as

$$P(d) = C(d) + a(d)[1 - C(d)] \quad (7)$$

where

$$a(d) = R_f \left(1 - \frac{d}{d_0}\right) \quad \text{for } d < d_0$$

$$a(d) = 0 \quad \text{for } d > d_0$$

As is clear from these equations, Del Villar and Finch also assumed that the experimental curve asymptotes to the water recovery at the finest particle sizes. Roldan-Villasana et al. [13] find that this later model is an improvement over that of Finch, particularly so in the modelling of coarse particle sizes. They argue that both these models suffer the disadvantage of not having a defined derivative and are able to describe only shallow fish-hooks.

An alternative is given by Hodouin et al. [14] who consider the feed as being split into two fractions. A fraction equal to the water recovery is classified by entrainment. This entrainment function, $C^*(d)$, has the same mathematical form as the classification function, $C(d)$. The coarse product of the entrainment is recombined with the remainder of the feed and classified by $C(d)$. The coarse product of the classification and the fine product of the entrainment functions are combined as the final product.

Mathematically, this is represented as

$$P(d) = R_f[1 - C(d)] + C^*(d)[R_f C(d) + (1 - R_f)] \quad (8)$$

This can be re-arranged as

$$P(d) = C(d) + R_f[1 - C^*(d)][1 - C(d)] \quad (9)$$

which shows that the assumption about the bypass equalling the water recovery is made, and that the second term consists of the product of two inverted partition functions.

Roldan-Villasana et al. [13] propose a model which subtracts a probabilistic entrainment effect from the corrected partition curve:

$$P(d) = (1 - R_f)C(d) + R_f - \frac{k_a d}{\exp(k_b d)} \quad (10)$$

The parameters k_a and k_b are indicative of the depth and breadth of the fish-hook, respectively. This model is shown to fit the data satisfactorily and the data are able to model the depth of the fish-hook as a function of cyclone inlet pressure. However, other data sets [15] indicate that negative values of the partition function can be obtained in certain instances. Furthermore, the probabilistic function used for the entrainment effect must be questioned as it indicates that smaller size particles are entrained to a lesser degree than larger sizes. It is argued that the smallest particle sizes act like the fluid, while somewhat larger particles will collide with larger particles and be entrained in their wake or will be trapped by them. This qualitative argument appears to be a justification for the functional form used.

3. A general partition curve model

It can be seen that the three models described [1,12,14] have a similar form that can be represented by

$$P(d) = C(d) + B_p[1 - E(d)] \quad (11)$$

Thus $P(d)$ can be interpreted as a combination of $C(d)$ and a dispersion function, $E(d)$, affecting a fraction, B_p , of the feed to the cyclone. Dispersion will be particle size dependent and can be expected to increase as the particles become smaller.

It is useful if the mathematical form of the dispersion function is chosen to be the same as that of the classification function. When the cutsize and sharpness of $E(d)$ are the same as for $C(d)$, the equation reverts to that for a partition curve without a fish-hook. If they are not equal, fish-hooks of any depth and breadth can be modelled.

If the dispersion and the classification functions are both modelled using the Plitt/Reid form, then the following expression is produced:

$$P(d) = 1 - \exp \left[-0.693 \left(\frac{d}{d_{50\text{class}}} \right)^{m_{\text{class}}} \right] + B_p \exp \left[-0.693 \left(\frac{d}{d_{50\text{disp}}} \right)^{m_{\text{disp}}} \right] \quad (12)$$

The classification is defined by the classification cutsize, $d_{50\text{class}}$, the classification sharpness, m_{class} , the dispersion by the dispersion cutsize, $d_{50\text{disp}}$ and the dispersion sharpness, m_{disp} . Note that in this general model, B_p is not assumed to

be equal to the water recovery. As previously noted, this assumption is not always valid, although it is, in many cases, a good approximation. Turbulent dispersion or other factors such as chemical dispersants [16] can affect the bypass. As will be shown later, the bypass can also be significantly larger than the water recovery, indicating that the conventional interpretation that an equivalent fraction of the feed solids follows the water to the underflow is not always correct.

3.1. Turbulence modelling of two-phase mixtures

Roco [17] describes a methodology for quantifying the relative importance of particle dispersion due to turbulence, and particle motion due to a gravitational field. These are referred to as dispersion and agglomeration forces (energy per unit time), respectively. In the context of hydrocyclone separation, the latter can be regarded as a classification force. The particles are dispersed in the carrier fluid if the dispersion due to the turbulence drag force is at least equal to the classification force due to the centrifugal acceleration.

Roco [17] defines a dispersion length scale, l_{dis} , for hydrodynamic, particulate two-phase flow. In a turbulent system, for instance, the turbulence mixing length is the dispersion length scale. A dispersion index, I_{dis} , can be defined as the ratio between the dispersion and the classification energy per unit time [17]. Since the classification energy can be expressed as a function of the classification force multiplied by the particle size, the dispersion index can be expressed as the ratio between l_{dis} and the particle size:

$$I_{\text{dis}} = \frac{l_{\text{dis}}}{d} \quad (13)$$

If I_{dis} is less than unity, particles will be classified due to the centrifugal force, otherwise (i.e. $I_{\text{dis}} > 1$), they will be dispersed within the fluid. If $I_{\text{dis}} > 10$, the dispersion can be regarded as quasi-homogenous.

For non-interacting solids, l_{dis} is given by [17]:

$$l_{\text{dis}} = \frac{(u^*)^2}{a(s - s_m)} \quad (14)$$

where u^* is the friction (or turbulent) velocity and s and s_m the relative densities of the solids and the suspension, respectively.

For a hydrocyclone, the centrifugal acceleration, a , is given by

$$a = \frac{v_i^2}{r_i} \quad (15)$$

where v_i is the tangential velocity at radius, r_i . The dispersion index, I_{dis} , at a given radius of the hydrocyclone and for a given particle size is, therefore, given by

$$I_{\text{dis}} = \frac{(u^*/v_i)^2 r_i}{d(s - s_m)} \quad (16)$$

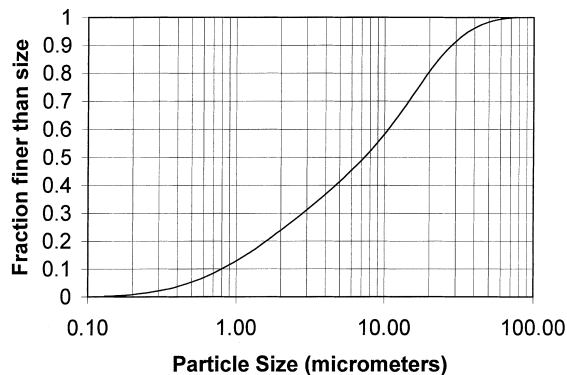


Fig. 1. Feed size distribution.

The estimated dispersion index can be compared to the observed separation of a particular particle size in a hydrocyclone, and if in reasonable agreement, it allows the performance to be attributed to the balance between the dispersion and the classification forces, as described in the general partition curve model.

4. Experimental method

A single 10 mm ceramic solid–liquid hydrocyclone supplied by Richard Mozley Ltd. (UK) was used for the experiments. The vortex finder diameter was 2.0 mm with a square inlet diameter of 1.7 mm. The spigot diameter was 1.0 mm. A Mono pump was used to feed the system and a bypass valve was used for pressure control.

The solid feed material was silica flour with a median size of 8.5 μm , supplied by Hepworth Minerals (UK). Fig. 1 shows the cumulative size distribution of the feed material. The feed concentration was 44 g l^{-1} for all the tests. The feed pressures used for this series of four experiments were 2.0, 3.0, 3.5 and 4.0 bar, respectively, with both of the outlets open to the atmosphere.

Samples were taken from the feed and simultaneously from the two product streams. A proportion was dried to determine the solids concentration. Overall mass balance discrepancies were less than 2%.

A sub-sample of each stream was size analyzed using a Malvern Mastersizer without drying to eliminate agglomeration. It was found, by comparing the experimentally measured solids recovery with the solids recovery estimated from the three fractional size analyses, that results below

0.6 μm were unreliable and were not used for modelling. In the region of the partition curve where the fish-hook was observed, this discrepancy was generally less than 5%, confirming the validity of the data.

The five parameters of the general equation (the classification and the dispersion cutoffs and sharpnesses, respectively, and the bypass) were simultaneously estimated by minimizing the sum-of-errors difference between the observed and the estimated partition values, while constraining the estimation to predict the experimentally measured solids recovery for the measured feed size distribution.

5. Results and discussion

5.1. Overall performance

The effect of increased feed pressure on the overall hydrocyclone performance is summarized in Table 1. As expected, the volumetric throughput, Q , and the recovery of solids to the underflow (total efficiency, R_s) both increase as the pressure increases. Since there is a simultaneous decrease in the recovery of water, R_f , the solids concentration of the underflow increases as the pressure increases. It can be noted from Table 1 that the total volumetric recovery, R_v , and the water recovery, R_f , are correlated, both decreasing as the pressure increases.

5.2. Classification performance

5.2.1. Partition curves

Fig. 2 shows the partition curves for feed pressures of 2, 3 and 4 bar (for clarity, the curve for 3.5 bar is not shown). It can be noted that the fish-hook effect is pronounced. Fig. 2 further shows that Eq. (12), very closely, describes the measured data.

5.2.2. Turbulence modelling

In order to calculate the dispersion index, an estimate of the turbulent velocity is required. Dyakowski and Williams [18] modelled turbulent flow in small diameter hydrocyclones using the Reynolds stress approach. The normal dimensionless component of the Reynolds stress tensor is equivalent to the ratio of the friction (or turbulent) velocity and the velocity component in the same direction, and can be used to estimate the dispersion index.

Table 1
Overall mass balance

Feed pressure (bar)	Throughput rate, Q (hr^{-1})	Solids recovery, R_s (%)	Water recovery, R_f (%)	Volumetric recovery, R_v (%)	Underflow concentration (g l^{-1})
2.0	155.90	72.91	16.56	19.04	168.8
3.0	187.27	76.15	15.45	18.00	178.1
3.5	197.01	77.65	15.24	17.90	185.4
4.0	212.95	78.09	14.80	17.39	184.5

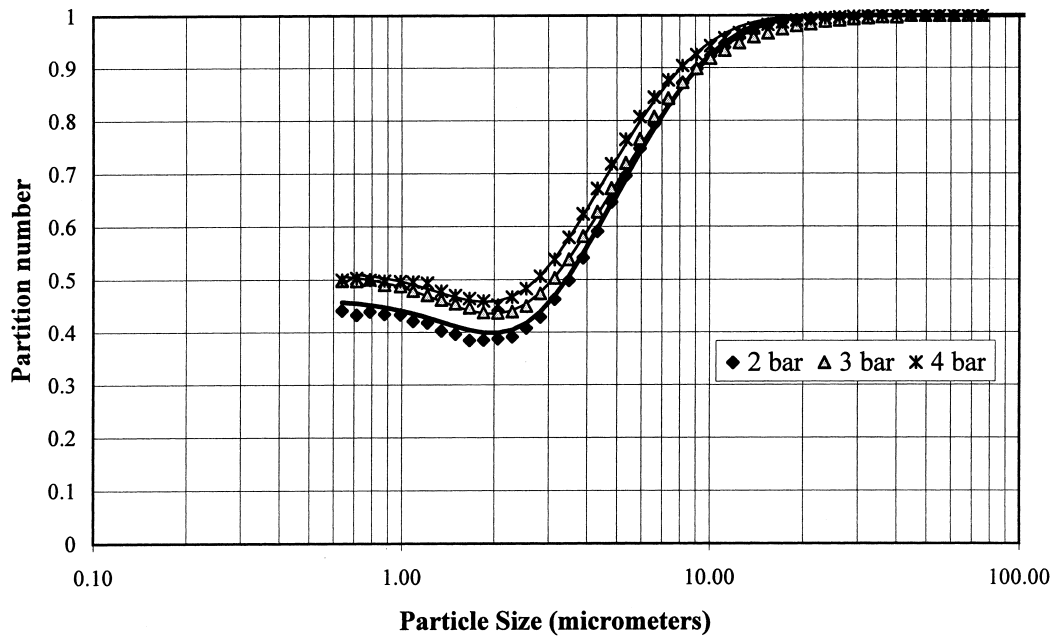


Fig. 2. Partition curves for 2, 3 and 4 bar feed pressure. Solid lines indicate the model fit.

Dyakowski and Williams calculated the dimensionless tangential velocity ratio (u^*/v_i) for small diameter cyclones of the same scale as used in this work, and found a linear increase from 0.10 to 0.175 between the center and the outside of the cyclone. Thus the dispersion index for different silica ($s = 2.7$) particle sizes as a function of radial position in the cyclone can be estimated using Eq. (16).

Fig. 3 shows the estimated dispersion index for particles of 1, 10 and 100 μm diameter. While these diameters span two orders of magnitude, the particle mass spans six orders of magnitude. Thus it is not unexpected that the dispersion and classification forces present in the hydrocyclone would affect the particles differently. It can be seen from Fig. 3 that for 100 μm particles, the dispersion index at all radial positions is less than 1, and this particle size will be

classified without significant dispersion. Particles of 10 μm diameter have a dispersion index less than 10, but generally greater than unity. This particle size, therefore, is influenced both by the dispersion and the classification forces, but is not expected to be quasi-homogeneously dispersed. Particles of 1 μm diameter are largely unaffected by the classification forces and can be considered to be quasi-homogeneously dispersed.

Comparisons of these results with the experimentally determined partition curves (Fig. 2) show close agreement. All 100 μm particles are classified to the underflow. 1 μm particles have partition values equal to the bypass value, i.e., complete dispersion. 10 μm particles are subject to a combination of dispersion and classification forces.

5.2.3. Bypass

Fig. 4 shows the estimated value of the bypass for the four tests. The bypass initially increases as the feed pressure increases, after which it appears to remain relatively con-

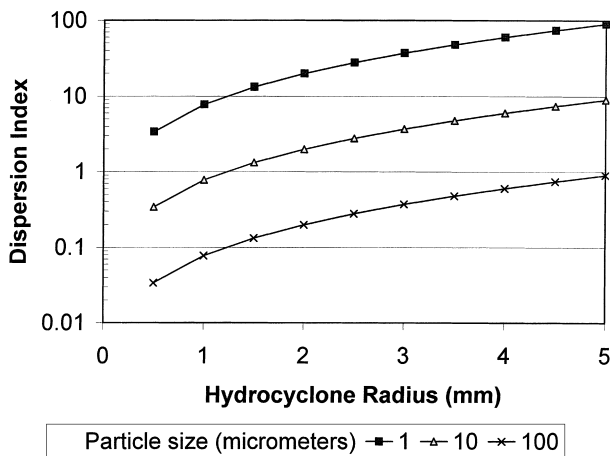


Fig. 3. Estimated dispersion index for 1, 10 and 100 μm particles.

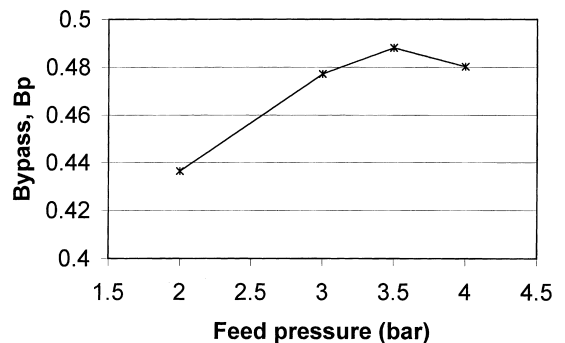


Fig. 4. The effect of feed pressure on the bypass.

Table 2
Classification parameters

Feed pressure (bar)	Classification cutsize, $d_{50\text{class}}$ (μm)	Dispersion cutsize, $d_{50\text{disp}}$ (μm)	Classification sharpness, m_{class}	Dispersion sharpness, m_{disp}
2.0	3.49	1.40	1.25	1.98
3.0	3.20	1.31	1.13	1.91
3.5	3.03	1.29	1.20	1.95
4.0	2.95	1.33	1.19	1.93

stant. There is some evidence that a further increase in pressure results in a subsequent decrease in the bypass, and this requires further investigation.

Both the lowest point of the fish-hook (Fig. 2) and the bypass (Fig. 4) are considerably higher than either the water recovery, R_f or the volumetric slurry recovery, R_v (Table 1). This phenomenon has previously not been observed to this degree, and it indicates that the assumption that the water recovery determines the bypass is not always correct.

It is possible that the observed bypass consists of two components: the dispersed solids recovered in proportion to water and an additional fraction, possibly due to boundary layer flow directly to the underflow. In small diameter cyclones, even a relatively thin boundary layer will comprise a significant proportion of the total volume of the hydrocyclone and will significantly increase the bypass. In larger diameter hydrocyclones, this fraction will be negligible and the bypass will, in general, correspond to the water recovery. Verification of this postulate is not currently possible as neither the boundary layer thickness nor the flow patterns in 10 mm hydrocyclones have been measured accurately.

5.2.4. Classification and dispersion

Table 2 summarizes the classification and the dispersion equation parameters. It can be seen that the classification cutsize decreases as the pressure increases. This is due to the higher centrifugal force produced at higher flow rates. The dispersion cutsize is not significantly affected by changes in feed pressure.

There does not appear to be a significant effect of pressure on the sharpness of either the classification or the dispersion functions.

6. Conclusions

A general mathematical form has been developed that describes satisfactorily the fish-hook partition curve. This equation can also be used to describe partition curves that do not display fish-hooks. The general equation consists of two competing effects: that of classification, described by a conventional corrected partition curve applied to the complete feed, and that of dispersion, described by an inverse corrected partition curve that is applied to only the bypass fraction. Turbulent two-phase modelling based on the

balance of dispersion to classification forces justifies the model.

Experimentation confirmed the fish-hook in small diameter hydrocyclones. The effect of pressure on the fish-hook was investigated. It was found that an increase in pressure led to a decrease in the classification cut-size, and an increase in the recovery, as expected. The classification sharpness and the dispersion cutsize and sharpness were unaffected. The bypass fraction increased as the pressure increased, increasing the fraction of the feed affected by the dispersive force.

The recovery of water to the underflow was significantly lower than either the lowest point of the partition curve at the bottom of the fish-hook or the value of the bypass. This shows that the assumption made frequently about the bypass equalling the water recovery is not generally applicable. It is possible that this is due to boundary layer flow directly to the underflow stream. Further work is required to conclusively determine the variation in the bypass with operating conditions.

7. Nomenclature

a	centrifugal acceleration (m s^{-2})
$a(d)$	size-dependent entrainment function
B_p	bypass
$C(d)$	corrected partition number
$C^*(d)$	entrainment function
d	particle size (m)
d_0	largest particle size affected by the fish-hook (m)
d_{50c}	corrected cut size (m)
$d_{50\text{disp}}$	dispersion cutsize (m)
$E(d)$	dispersion function
$f(d)$	mass fraction of particle size d in the hydrocyclone feed
F	mass flow rate of solids in hydrocyclone feed (kg s^{-1})
I_{dis}	dispersion index
k_a, k_b	parameters determining the depth and breadth of the fish-hook
l_{dis}	dispersion length scale for hydrodynamic, particulate two-phase flow (m)
m	parameter indicating the sharpness of separation
m_{class}	classification sharpness parameter
m_{disp}	dispersion sharpness parameter

$P(d)$	partition number
Q	volumetric throughput ($\text{m}^3 \text{s}^{-1}$)
r_i	radial position in hydrocyclone (m)
R_f	fractional water recovery from feed to underflow
R_s	fractional recovery of solids to the underflow (total efficiency)
R_v	fractional volumetric recovery from feed to underflow
s, s_m	relative densities of solids and suspension (kg m^{-3})
U	mass flow rate of solids in hydrocyclone underflow (kg s^{-1})
$u(d)$	mass fraction of particle size d in the hydrocyclone underflow
u^*	friction (or turbulent) velocity (m s^{-1})
v_i	tangential velocity (m s^{-1})

References

- [1] J.A. Finch, Modelling a fish-hook in hydrocyclone selectivity curves, *Powder Technol.* 36 (1983) 127–129.
- [2] N. Yoshioka, Y. Hotta, Liquid cyclone as a hydraulic classifier, *Chem. Eng. Jpn (Transl.)* 19 (1955) 632–640.
- [3] L.R. Plitt, A mathematical model of the hydrocyclone classifier, *CIM Bull.* (1976) 114–122.
- [4] K.J. Reid, Derivation of an equation for classifier performance curves, *Can. Metall. Q.* 10(3) (1971) 253–254.
- [5] A.J. Lynch, T.C. Rao, Modelling and scale-up of hydrocyclone classifiers, in: *Proc. 11th Int. Mineral Processing Cong., Cagliari*, 1975, pp. 245–269.
- [6] B.C. Flinthoff, L.R. Plitt, A.A. Turak, Cyclone modelling: a review of present technologies, *CIM Bull.* (1987) 39–50.
- [7] L.G. Austin, R.R. Klimpel, P.T. Luckie, *Process Engineering of Size Reduction: Ball Milling*, SME of American Institute of Mining, Metallurgical and Petroleum Engineers, New York, 1984.
- [8] A.J. Lynch, *Mineral Crushing and Grinding Circuits: Their Simulation, Optimization, Design and Control*, Elsevier, Amsterdam, 1977, 340 pp.
- [9] L.G. Austin, R.R. Klimpel, An improved method for analyzing classifier data, *Powder Technol.* 29 (1981) 277–281.
- [10] L. Svarovsky, *Hydrocyclones*, Holt, Rinehart and Winston, London, 1992, 198 pp.
- [11] T. Braun, M. Bohnet, Influence of feed solid concentration on the performance of hydrocyclones, *Chem. Eng. Technol.* 13 (1990) 15–20.
- [12] R. Del Villar, J.A. Finch, Modelling the cyclone performance with a size dependant entrainment factor, *Miner. Eng.* 5(6) (1992) 661–669.
- [13] E.J. Roldan-Villasana, R.A. Williams, T. Dyakowski, The origin of the fish-hook effect in hydrocyclone separators, *Powder Technol.* 77 (1993) 243–250.
- [14] D. Hodouin, S. Caron, J.J. Grand, Modelling and simulation of a hydrocyclone desliming unit, in: *Proc. 1st World Cong. on Particle Technology, Nurnberg, Part IV*, 1986, pp. 507–522.
- [15] E.J. Roldan-Villasana, Modelling and simulation of hydrocyclone networks for fine particle processing, Ph.D. Thesis, UMIST, Manchester, UK, 1992.
- [16] R.R. Klimpel, The influence of chemical dispersant on the sizing performance of a 24-in hydrocyclone, *Powder Technol.* 31 (1982) 255–262.
- [17] M.C. Roco, One equation turbulence modelling of incompressible mixtures, in: N.P. Cheremisinoff (Ed.), *Encyclopedia of Fluid Mechanics*, vol. 10, 1996, pp. 1–68.
- [18] T. Dyakowski, R.A. Williams, Modelling turbulent flow within a small-diameter hydrocyclone, *Chem. Eng. Sci.* 48 (1993) 1143–1152.