

PREDICTION OF THE EXTREMAL SHAPE FACTOR OF SPHEROIDAL PARTICLES

DANIEL HLUBINKA AND VIKTOR BENEŠ

Department of Probability and Statistics, Charles University, Sokolovská 83, 186 75 Praha 8, Czech Republic
 E-mail: hlubinka@karlin.mff.cuni.cz

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ABSTRACT

In the stereological unfolding problem for spheroidal particles the extremal shape factor is predicted. The theory of extreme values has been used to show that extremes of the planar shape factor of particle sections tend to the same limit distribution as extremes of the original shape factor for both the conditional and marginal distribution. Attention is then paid to the extreme shape factor conditioned by the particle size. Normalizing constants are evaluated for a parametric model and the numerical procedure is tested on real data from metallography.

Keywords: extremal shape factor, spheroidal particles, unfolding.

INTRODUCTION

Consider a random collection of spheroidal particles in a reference volume. We shall assume that the particles are all oblate or all prolate. The size is the length of a particle's major semiaxis X , while W is the length of the minor semiaxis. The shape factor in our setting is given by $T = X^2/W^2 - 1$. It is clear that $0 < W \leq X < \eta$, and $0 \leq T < \omega$, where the equalities $X = W$ and $T = 0$ hold when the particles are balls. Both X and W are nonnegative real random variables. Values η and ω are fixed non-negative real numbers, possibly infinite, the upper end-points of the supports of distribution of X and T respectively. We study random planar sections of the particles. These sections form ellipses. An ellipse is characterized by its size (major semiaxis length) Y , and its shape factor $Z = Y^2/V^2 - 1$, where V is the length of the minor semiaxis. It holds that $0 < Y \leq X$, and $0 \leq Z \leq T$.

Let $g(x, t)$ be the joint probability density function of size and shape factor (X, T) of a particle. The orientation of the particle is assumed to be isotropic, i.e. a uniform random variable on the hemisphere independent of size and shape. On the other hand we do not assume independence between size and shape factor.

Following (Cruz-Orive, 1976) the distribution of the section size and shape factor (Y, Z) for oblate particles has the joint probability density

$$f(y, z) = \frac{y\sqrt{1+z}}{2M} \int_y^\eta \int_z^\omega \frac{g(x, t) dt dx}{\sqrt{t}\sqrt{1+t}\sqrt{t-z}\sqrt{x^2-y^2}}, \quad (1)$$

where $2M$ is the population mean caliper diameter of particles. We will need the joint distribution of the

original size X and planar shape factor Z as well. Using similar arguments we get the probability density

$$h(x, z) = \frac{\sqrt{1+z}}{M_x} \int_z^\omega \frac{g(x, t) dt}{\sqrt{t}\sqrt{1+t}\sqrt{t-z}}, \quad (2)$$

where

$$\begin{aligned} M_x &= \int_0^\omega \frac{g_x(t)}{\sqrt{t(1+t)}} \left(\int_0^t \sqrt{\frac{z+1}{t-z}} dz \right) dt \\ &= \int_0^\omega \left[(t+1)^{-\frac{1}{2}} + \sqrt{\frac{t+1}{t}} \arctan \sqrt{t} \right] g_x(t) dt, \end{aligned}$$

and $g_x(t) = g(x, t) / \int_z^\omega g(x, t) dt$ is the conditional density of T given $X = x$. Then the complementary equation which together with (2) yields (1) is

$$f(y, z) = \frac{y}{2M} \int_y^\eta \frac{M_x h(x, z)}{\sqrt{x^2-y^2}} dx. \quad (3)$$

We denote $f_y(z)$ the conditional density of shape factor Z given size $Y = y$. The conditional probability density of Z given $X = x$ is equal to

$$h_x(z) = \frac{\sqrt{1+z}}{M_x} \int_z^\omega \frac{g_x(t) dt}{\sqrt{t}\sqrt{1+t}\sqrt{t-z}}. \quad (4)$$

Further let $h(z)$ denote the marginal density of the transformed shape factor Z . We have

$$h(z) = \sqrt{1+z} \int_z^\omega \frac{1}{\sqrt{t}\sqrt{1+t}\sqrt{t-z}} \int_0^\eta \frac{g(x, t)}{M_x} dx dt.$$

Finally define $f_z(y)$, $h_z(x)$ as the conditional density of size Y given shape $Z = z$, and size X given shape $Z = z$, respectively.

For the sake of completeness we provide the formula for the density of the section characteristics for prolate particles which is expressed in terms of shorter semiaxes

$$f(v, z) = \frac{v}{2M(1+z)^2} \int_v^\infty \int_z^\infty \frac{(1+t)^{3/2} g(w, t) dt dw}{\sqrt{t}\sqrt{t-z}\sqrt{w^2-v^2}}.$$

In the following, however, we restrict consideration to oblate particles. For the prolate particle case analogous results hold.

EXTREMAL PROPERTIES

A univariate distribution function K is said to belong to the domain of attraction of a distribution function L if there exist normalizing constants $\{a_n\}$, $\{b_n\}$ such that the n -th power $K^n(a_n x + b_n)$ converges weakly to L , where L is one of the following distributions

$$L_{i,\gamma}(x) = \begin{cases} \exp(-x^{-\gamma}), & x \geq 0, \quad i = 1, \quad \text{Fréchet} \\ \exp(-(-x)^{-\gamma}), & x \leq 0, \quad i = 2, \quad \text{Weibull} \\ \exp(-e^{-x}), & x \in \mathbb{R}, \quad i = 3, \quad \text{Gumbel} \end{cases} \quad (5)$$

where $\gamma > 0$. We shall write $K \in \mathcal{D}(L)$ if K is in the domain of attraction of L .

If the distribution K has a density k , there are sufficient conditions (de Haan, 1975) for K to be in $\mathcal{D}(L)$. These conditions are denoted $(C_{1,\gamma})$, $(C_{2,\gamma})$, (C_3) .

In Drees and Reiss (1992) the extremal properties in the Wicksell corpuscule problem were studied. This was developed so as to apply to the size-shape unfolding problem of spheroidal particles. Let H_x , H , G_x , F_y , F_z , H_z be the distribution functions corresponding to densities h_x , h , g_x , f_y , f_z , h_z respectively. Then the following theorem holds (Hlubinka, 2000):

Theorem 1 a) Suppose that for any fixed size x the density $g_x(t)$ satisfies condition $C_{i,\gamma}$. Then the distribution $H_x \in \mathcal{D}(L_{i,\beta})$, $i = 1, 2, 3$, where $\beta = \gamma$ for $i = 1$, and $\beta = \gamma + 1/2$ for $i = 2$.

b) Assume that $g_x(t)$ satisfies condition $C_{i,\gamma}$ uniformly in x . Then $F_y \in \mathcal{D}(L_{i,\beta})$ for all y , $i = 1, 2, 3$, where $\beta = \gamma$ for $i = 1$, and $\beta = \gamma + 1/2$ for $i = 2$.

c) Assume that $g_x(t)$ satisfies condition $C_{i,\gamma}$ uniformly in x . Then $H \in \mathcal{D}(L_{i,\beta})$, $i = 1, 2, 3$, where $\beta = \gamma$ for $i = 1$, and $\beta = \gamma + 1/2$ for $i = 2$.

The theorem says that the domain of attraction for the shape factor is the same for a particle and its section. We will not study further part c) since it is less important for practical applications. Indeed, the total extremal shape factor is useless if it is not related to particle size.

To use the results of theorem 1, part a), we need to estimate normalizing constants a_n, b_n, a'_n, b'_n such that conditioned by the known particle's size $X = x$

$$\frac{T_{(n)} - b_n}{a_n} \xrightarrow{\mathcal{D}} \Lambda, \\ \frac{Z_{(n)} - b'_n}{a'_n} \xrightarrow{\mathcal{D}} \Lambda,$$

where $T_{(n)}$ is the maximum of n observations, and Λ is a random variable with the extremal type of distribution.

A parametric model of gamma distribution will be considered with the density

$$g_x(t; \alpha, \mu) = \frac{t^{\alpha-1} \mu^\alpha}{\Gamma(\alpha)} e^{-\mu t}, \quad t \geq 0. \quad (6)$$

where α and μ are positive constants which can possibly depend on the given size x . Recall that the gamma distribution is in the domain of attraction of the Gumbel distribution. Using the techniques of Takahashi (1987) the following theorem was proved in Hlubinka (2000):

Theorem 2 Assume that for any fixed size $X = x$ the shape factor T follows a gamma distribution. Then both T and Z conditioned on $X = x$ belong to the Gumbel domain of attraction and their normalizing constants are

$$a_n = \frac{1}{\mu}, \\ b_n = \frac{1}{\mu} \left[\log n + (\alpha - 1) \log \log n + \log \frac{1}{\Gamma(\alpha)} \right], \\ a'_n = \frac{1}{\mu}, \\ b'_n = \frac{1}{\mu} \left[\log n + \left(\alpha - \frac{3}{2} \right) \log \log n + \log \frac{\sqrt{\pi}}{M_x \Gamma(\alpha)} \right].$$

Using Theorem 2 we can approximate the distribution $G_x^n(t)$ of $T_{(n)}$ by $\Lambda((t - b_n)/a_n)$. Quantiles of $T_{(n)}$ are estimated via $\hat{q}_p = b_n + a_n[-\log(-\log p)]$, and $\widehat{ET}_{(n)} = b_n + a_n C$, where $C \doteq 0.5772$ is the Euler constant. Concerning the estimation of normalizing constants a_n, b_n using estimation of a'_n, b'_n , several methods concerning the corpuscule problem are

discussed in Takahashi and Sibuya (1998). We can use for example a maximum likelihood method based on the joint distribution of $(T_{(n-k+1)}, \dots, T_{(n)})$ which is derived in Weissman (1968) to estimate a'_n and b'_n . Note that since $a'_n = a_n = 1/\mu$ does not depend on n the main problem is to find a reasonable estimation of b_n based on $\widehat{1/\mu}$ and \widehat{b}'_n . This means the use of numerical methods since α is unknown.

For the situation b) from theorem 1, unfortunately, such a simple result as Theorem 2 is not available.

PRACTICAL APPLICATION

The prediction of the extremal particle shape factor is important in metallography. Engineers claim that damage to construction materials depends on extremal rather than mean characteristics of the microstructure. Therefore corresponding methods for prediction of spatial extremes based on data from sections (metallographic samples) have to be developed. In Beneš *et al.* (1997) the problem of unfolding the size-shape-orientation distribution has been theoretically solved. We use data from that study, i.e. measurement of an aluminium alloy specimens with oblate Si particles. The assumptions of spheroidal shape and isotropy are approximately fulfilled. Metallographic samples were measured by means of an image analyser to obtain samples from the joint distribution with density $f(y, z)$. The aim is to use Theorems 1 and 2 for the prediction of the extremal shape factor of particles for given size classes.

The gamma distribution parametric model (6) for the shape factor given the size is used. Our approach is to use formula (3) for unfolding the size distribution of particles to obtain an empirical distribution corresponding to the density $h(x, z)$. This is then fitted to the theoretical distribution obtained by plugging (6) into (4). This serves two purposes: first we can evaluate the degree of fit and conclude whether the model is appropriate, secondly the parameters α and μ are estimated and normalizing constants a_n, b_n obtained from Theorem 2.

In the first step we use discretization and the EM-algorithm for the unfolding as described generally e.g. in Ohser and Mücklich (2000). Equation (3) is transformed to:

$$1 - F_z(y) = \frac{1}{2M} \int_y^\eta p(x, y) h_z(x) dx, \quad (7)$$

where

$$p(x, y) = M_x \sqrt{x^2 - y^2}.$$

It holds that

$$2M = \frac{N_A}{N_V},$$

where N_V is the mean number of particles per unit volume and N_A is the mean number of particle sections per unit area of the section plane. Equation (7) is thus transformed to

$$N_A(1 - F_z(y)) = N_V \int_y^\eta p(x, y) h_z(x) dx, \quad (8)$$

where N_A is estimated from the data. The y variable is subdivided into classes with endpoints $y_k = b^k$, b a given constant. Then for $\zeta_k = N_A(F_z(y_k) - F_z(y_{k-1}))$ we have

$$\zeta_k = N_V \int_y^\eta [p(x, y_{k-1}) - p(x, y_k)] h_z(x) dx.$$

Assume that $H_z(x)$ can be discretized to have values a_i for $x_{i-1} \leq x < x_i$, where $x_i = b^i$, then

$$\zeta_k = N_V \sum_i p_{ik}(a_i - a_{i-1}) = \sum_i p_{ik} \xi_i,$$

where $p_{ik} = p(x_i, y_{k-1}) - p(x_i, y_k)$, $i \geq k$, $p_{ik} = 0$ otherwise. It holds that $\xi_i = N_V(a_i - a_{i-1})$, from which the estimator of N_V is $N_V = \sum_i \xi_i$. Now the iteration of the EM-algorithm is given by $(v + 1)$ -step:

$$\xi_i^{v+1} = \frac{\xi_i^v}{q_i} \sum_{k=1}^i \frac{p_{ik}}{r_k} \zeta_k, \quad (9)$$

where ξ_i^v is the v -th iterated ξ_i , $q_i = \sum_k p_{ik}$, $r_k = \sum_{i \geq k} p_{ik} \xi_i^v$, and the initial value ξ_i^0 can be taken to be ζ_j . In this way probabilities ξ_i/N_V of the discrete conditional distribution $H_z(x)$ given z are estimated. Using the empirical distribution function $H(z) = F(z)$, also the contrary conditional distribution function $H_x(z)$ can be directly estimated from $H_z(x)$.

Then for the shape factor subintervals $(z_{i-1}, z_i]$ denote $H_j^{\text{em}} = H_x(z_i) - H_x(z_{i-1})$ for fixed x . The corresponding theoretical probabilities H_j^{th} for the gamma model are obtained from (4) and (6) as

$$H_j^{\text{th}} = \int_{z_{j-1}}^{z_j} h_x(z) dz = \frac{1}{M_x} \frac{\mu^\alpha}{\Gamma(\alpha)} [I_1 + I_2],$$

where

$$I_1 = \int_{z_{j-1}}^{z_j} \frac{t^{\alpha-1} e^{-\mu t}}{\sqrt{t(1+t)}} B_t(z_{j-1}, t) dt,$$

$$I_2 = \int_{z_j}^\infty \frac{t^{\alpha-1} e^{-\mu t}}{\sqrt{t(1+t)}} B_t(z_{j-1}, z_j) dt,$$

and the integral $B_t(s, u) = \int_s^u \sqrt{(1+z)/(t-z)} dz$ is given in the expression for M_x (following (2)). Since in fact the unknown parameters play a role already in the unfolding step (through the M_x term in kernel function p), it is useful to suggest an analytical function modelling the dependence of the parameters μ, α on x .

For a given size class we estimate the parameters μ, α by minimizing either the $\sum_i (H_i^{\text{th}} - H_i^{\text{em}})^2$ or $\max_j |\sum_{i \leq j} H_i^{\text{th}} - \sum_{i \leq j} H_i^{\text{em}}|$.

NUMERICAL RESULTS

A sample of $m = 5694$ particle sections of the aluminium alloy discussed in Beneš *et al.* (1997) was evaluated using the procedure described in the previous sections. Si particles are oblate and there is a strong correlation between the shape factor and size: larger particles are thin with large shape factor and vice versa. From the total measured area A we obtain the estimator $N_A = n/A = 0.00125 \mu\text{m}^{-2}$. Using the unfolding procedure we get $N_V = 2.1 \times 10^{-4} \mu\text{m}^{-3}$.

Consider the subdivision into 4×4 classes of bivariate size-shape histogram with size class factor $b = 2.99 \mu\text{m}$. The upper bounds of intervals x are in the Table 1 below together with relative frequencies $f(x)$ of the unfolded size distribution. All

size characteristics are in micrometers. Estimation of parameters was done under the model of α fixed and $\mu = c_1 x^{c_2}$. Using the least squares criterion we obtained $\alpha = 1.41$, $c_1 = 1.40$, $c_2 = -1.356$. The estimated values μ, α are in the Table. Further we choose n for each size class corresponding to the relative frequency $g(x)$ in the unfolded sample. The normalizing constants a_n, b_n are obtained from Theorem 2 and finally the extremal shape factor characteristics $\widehat{ET}_{(n)}, \hat{q}_{0.95}$ for given n .

These numerical results demonstrate the usefulness of the theory outlined above. For comparison the largest observed particle section shape factor in the particle population investigated was 1808.6 (corresponding to the last size class of the largest particles). The results may be improved by a more detailed model for parameters (e.g. non-constant α) which would make the numerical solution more difficult. Also the analysis may be performed for a larger number of classes. The open problem remains still the application of b) in Theorem 1, which would be more straightforward.

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Table 1. *Numerical results.*

x	$g(x)$	α	μ	n	a_n	b_n	$\widehat{ET}_{(n)}$	$\hat{q}_{0.95}$
2.99	0.22	1.41	0.317	1253	3.15	25.4	27.2	34.8
8.92	0.48	1.41	0.072	2733	13.89	123.1	131.0	164.4
26.7	0.26	1.41	0.0163	1480	61.35	504.3	539.4	686.5
79.7	0.04	1.41	0.0037	228	270.27	1683.2	1837.8	2486.0

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