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Correlations between fragment sizes in sequential fragmentation

Mikko Lensu†

Ship Laboratory, Helsinki University of Technology, Tietotie 1, FIN-02150 Espoo, Finland

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Abstract. Size correlations in a sequential fragmentation process originate from the correlations the progeny of a single fragmentation event necessarily have. A multidimensional equation describing the binary fragmentation of a unit segment is formulated. The fragments are assumed to keep their positions with respect to the original segment. If the fragmentation rate is not dependent on fragment size the distribution $f_n(x_1, x_2, \ldots, x_n)$ for n successive adjacent fragments is shown to asymptotically approach an n-dimensional log-normal distribution. Explicit forms of the asymptotic distribution and the correlations are given in terms of the parameters of $B(y \to x)$, the probability that a fragmentation event with parent size y produces progeny size x.

1. Introduction

Fragmentation processes are ubiquitous in physics and are encountered in such fields as dispersion, turbulent mixing, polymer chemistry, crushing and grinding of minerals, aerosol behaviour, nuclear physics and astronomy (Ramkrishna 1985, Mekijan and Lee 1991). The associated linear fragmentation equation has been extensively studied and various analytical solutions have been obtained (McGrady and Ziff 1987, Ziff 1991, Hassan and Rodgers 1995, Cheng and Redner 1990). For a fragment size independent fragmentation rate the solutions are known or suspected to approach asymptotically a lognormal distribution (Epstein 1947, Delannay *et al* 1996, Baker *et al* 1992). The log-normality is expected to be largely independent of the form of the probability $B(y \to x)$ for producing x-fragments from y-fragments. This makes the lognormal distribution particularly suitable for modelling empirical fragment size distributions.

If there is no mass loss then in a fragmentation event the sizes of the progeny fragments are necessarily correlated as their sum must equal the size of the parent fragment. For binary fragmentation the correlations are deterministic as the sizes of the progeny for a parent size y are x and $y - x$. If the fragments are free to move around the correlations have little relevance. However, if they have fixed positions, a specific geometry or cellular structure is generated and correlations between a fragment and its neighbours are expected. This situation is also often encountered in fragmentation phenomena, especially in the fragmentation of solids where generation of crack surfaces and the associated change in material properties are often in question.

† E-mail address: Mikko.Lensu@hut.fi

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2. The distribution of adjacent fragments

2.1. Preliminaries

The model to be presented generalizes the usual population balance or rate equation approach to describe arrangements of adjacent fragments. This is done in an iterative fashion starting from a theory of two adjacent fragments. An unit segment is fragmented sequentially into $N(t)$ fragments that are assumed to keep their positions. The distribution of a random vector (X, Y) where X and Y are the sizes of adjacent fragments is $f(x, y)\,\mathrm{d}x\,\mathrm{d}y = \mathrm{Prob}(x \leqslant X < x + \mathrm{d}x$ and $y \leqslant Y < y + \mathrm{d}y)$. The distribution is assumed to by symmetric so that the ordinary fragment size distribution is $f(x) = \int_0^1 \mathrm{d}y\, f(x, y)$ and the conditional distribution

$$f(y|X = x) = \frac{f(x, y)}{f(x)}. \tag{1}$$

The distribution $f_n(x_1, x_2, \ldots, x_n)$ for n successive adjacent fragments is defined as an obvious generalization. The population balance equation for the distribution $f(x)$ is

$$\frac{\partial}{\partial t} N f(x) = N2 \int_x^1 \mathrm{d}z\, \alpha(z) f(z) B(z \to x) - N\alpha(x) f(x) \tag{2}$$

where $\alpha(x, t)$ is the fragmentation rate and $B(z \to x)\,\mathrm{d}x$ is the probability for a fragmentation event with parent size z to produce progeny size x. It is assumed that $\alpha = 1$ and that the fragmentation pattern is size independent or $B(z \to x)\,\mathrm{d}x \equiv B(x/z)(\mathrm{d}x/z)$. Here $B(y)\,\mathrm{d}y$ is a probability distribution defined for $0 \leqslant y \leqslant 1$. Applying to (2) the transform

$$G(p) = \int_0^1 \mathrm{d}x\, x^p f(x) \qquad D(p) = \int_0^1 \mathrm{d}x\, x^p B(x) \tag{3}$$

one obtains

$$\frac{\partial}{\partial t} N G(p) = N2G(p)D(p) - G(p) = NG(p)(2K(p) + 1) \tag{4}$$

where $K(p) = D(p) - 1$. It is seen that $G(p)$ is the Laplace transform of the distribution of $\ln(1/X)$ or the moment generating function of the distribution of $\ln(X)$; thus a convolution property derivable from that of Laplace transforms applies. With $G(0) = 1$ and $K(0) = 0$ two equations are obtained from (4),

$$\frac{\partial}{\partial t} G(p) = 2K(p)G(p) \qquad \frac{\mathrm{d}N}{\mathrm{d}t} = N \tag{5}$$

and the solution for any initial distribution $f_0(x)$ with the transform $G_0(p)$ at $t = 0$ is

$$G(p) = G_0(p)\, \mathrm{e}^{2K(p)t} \qquad N = N_0\, \mathrm{e}^t. \tag{6}$$

It can be assumed that $G_0(p) = 1$ and $N_0 = 1$ since the complete solution can always be derived as a convolution integral. The solution $f(x)$ for any $B(x)$ can formally be written down as a series where the kth term contains a k-fold convolution integral; for certain choices of $B(x)$ the series reduces to known functions (McGrady and Ziff 1987).

For the asymptotic solution consider the distribution of $\ln(X)$. The expectation of $\ln(X)$ is $\ln\mu$, where μ is the geometric mean of X, and the variance of $\ln(X)$ is $\ln^2\sigma$, where σ is the geometric standard deviation of X. These are obtained from $G(p)$ as

$$\ln\mu = \frac{\partial}{\partial p} \ln G(p)|_{p=0} = 2L_1 t \qquad \ln^2\sigma = \frac{\partial^2}{\partial p^2} \ln G(p)|_{p=0} = 2L_2 t \tag{7}$$

where L_n are the logarithmic moments of $B(x)$,

$$L_n = \int_0^1 \mathrm{d}x \ln^n x \, B(x) = \frac{\partial^n}{\partial p^n} K(p)|_{p=0} \qquad n \geqslant 1. \tag{8}$$

By expressing $K(p)$ in (6) as a series of L_n, transforming to a normalized variable and obtaining the limit of the moment generating function for $t \to \infty$ it is seen that the distribution of $\ln(X)$ is asymptotically normal $N(\ln\mu, \ln^2\sigma)$. Consequently $f(x)$ is asymptotically lognormal with geometric mean μ and geometric standard deviation σ,

$$f_{\mathrm{as}}(x) = \frac{1}{\ln\sigma\sqrt{2\pi}} \frac{1}{x} \exp\left\{ -\frac{1}{2}\left(\frac{\ln(x/\mu)}{\ln\sigma}\right)^2 \right\}. \tag{9}$$

With variable normalization it is also possible, at least for certain special cases, to derive an asymptotic form of (2) having (9) as solution. It follows that the multidimensional distributions pertaining to the process are asymptotically multidimensional lognormal distributions; their explicit form is derived below. It is probable that the condition imposed on $B(z \to x)$ is not necessary and that the lognormal asymptotics holds more generally for $\alpha = 1$. Cheng and Redner (1990) also obtained lognormal small fragment behaviour for certain scaling solutions with $\alpha(x) \sim x^\lambda$, $\lambda > 0$; for this case the scaling equation can be identified as the asymptotic form of (2).

2.2. *Evolution equation for the distribution of two adjacent fragments*

It is assumed that the Y-fragment is always on the same side of the X-fragment so that the number of fragments equals that of the fragment pairs for large N (strictly: is $N+1$). The equation governing the time change of the fragment pair distribution $N(t)f(x, y; t)$ is

$$\begin{aligned} \frac{\partial}{\partial t} N f(x, y) = {} & N \int_x^1 \mathrm{d}z \, \alpha(z) f(z, y) B(z \to x) \\ & + N \int_y^1 \mathrm{d}z \, \alpha(z) f(x, z) B(z \to y) + N\alpha(x+y) f(x+y) B(x+y \to y) \\ & - N(\alpha(x) + \alpha(y)) f(x, y) \end{aligned} \tag{10}$$

where the terms on the right-hand side are: the rate of creation of (x, y)-pairs from (z, y)-pairs, the rate of creation of (x, y)-pairs from (x, z)-pairs, the rate or creation of (x, y)-pairs from $(x+y)$-fragments, and the rate of annihilation of (x, y)-pairs.

2.3. *Asymptotic solution for $\alpha = 1$*

Assume again that $\alpha = 1$, $B(z \to x)\,\mathrm{d}x \equiv B(x/z)(\mathrm{d}x/z)$, and define the two-dimensional transform

$$G(p, q) = \int_0^1 \mathrm{d}x \, x^p \int_0^1 \mathrm{d}y \, y^q f(x, y). \tag{11}$$

The left-hand side and the last term on the right-hand side of (10) transform directly. Applying to the first term on the right-hand side and using the convolution property and (1) produces $NG(p, q)D(p)$. The second term is transformed similarly. Taking into account that $x + y \leqslant 1$ must apply, the third term is developed as

$$\begin{aligned} & N \int_0^1 \mathrm{d}x \, x^p \int_0^{1-x} \mathrm{d}y \, y^q f(x+y) B\left(\frac{x}{x+y}\right) \frac{1}{x+y} \\ & \quad = N \int_0^1 \mathrm{d}x \, x^{p+q} \int_x^1 \mathrm{d}y \left(\frac{y}{x} - 1\right)^q f(y) B\left(\frac{x}{y}\right) \frac{1}{y} = NG(p+q)E(p, q) \end{aligned} \tag{12}$$

where

$$E(p,q)=\int_0^1 \mathrm{d}x\, x^p(1-x)^q B(x). \tag{13}$$

The equation for the transform $G(p,q)$ is

$$\frac{\partial}{\partial t}NG(p,q)=N(K(p)+K(q))G(p,q)+NG(p+q)E(p,q). \tag{14}$$

Taking into account $G(p+q)=\exp(2K(p+q)t)$ and $N=\exp(t)$, (6), the solution satisfying the necessary condition $G(0,0)=1$ is

$$G(p,q)=\frac{E(p,q)\exp\{2K(p+q)t\}}{2K(p+q)-K(p)-K(q)+1}. \tag{15}$$

For the derivation of the lognormal limit distribution write (15) first as a single exponential. The terms in the exponent are expanded as a series of the L_n, (8):

$$\begin{aligned}
&\ln E(p,q)=1+(p+q)L_1+\tfrac{1}{2}(p^2+q^2)(L_2-L_1^2)+pq(\varepsilon-L_1^2)+\cdots\\
&\ln(K(p+q)-K(p)-K(q)+1)=1+(p+q)L_1+\tfrac{1}{2}(p^2+q^2)(L_2-L_1^2)\\
&\qquad+pq(2L_2-L_1^2)+\cdots\\
&2tK(p+q)=2t((p+q)L_1+\tfrac{1}{2}(p+q)^2L_2+\cdots).
\end{aligned} \tag{16}$$

Here

$$\varepsilon=\int_0^1 \mathrm{d}x\,\ln(x)\ln(1-x)B(x). \tag{17}$$

Insertion to (18) gives

$$G(p,q)=\exp\{pq(\varepsilon-2L_2)+2t(p+q)L_1+2t(p+q)^2L_2+\cdots\} \tag{18}$$

and changing to normalized variables

$$\ln x^*=\frac{\ln x-\ln\mu}{\ln\sigma}=\frac{\ln x-2L_1t}{\sqrt{2L_2t}} \qquad \ln y^*=\frac{\ln y-2L_1t}{\sqrt{2L_2t}} \tag{19}$$

changes the transform to

$$\begin{aligned}
G^*(p,q)&=\exp\left\{\frac{2L_1t}{\sqrt{2L_2t}}(p+q)\right\}G\left(\frac{p}{\sqrt{2L_2t}},\frac{q}{\sqrt{2L_2t}}\right)\\
&=\exp\left\{\frac{1}{2}(p^2+q^2+2\rho pq)+\mathrm{O}\left(\frac{1}{\sqrt{t}}\right)\right\}.
\end{aligned} \tag{20}$$

Taking the limit $t\to\infty$ one obtains the transform of $N(\ln x,\ln y,0,1;\rho)$, the symmetric bivariate normal distribution with correlation ρ and marginal distribution expectation and standard deviation 0 and 1, respectively. This establishes the result. The correlation is explicitly

$$\rho=\frac{\varepsilon+2L_2(t-1)}{2L_2t}. \tag{21}$$

That this is identical to the correlation of the exact solution $f(x,y)$ of (11) can be checked from (15) by the definition

$$\rho=\frac{E(\ln x\ln y)-\ln^2\mu}{\ln^2\sigma}=\frac{1}{\ln^2\sigma}\frac{\partial^2}{\partial p\partial q}\ln G(p,q)|_{p=q=0}. \tag{22}$$

Scaling back by (17), the asymptotic distribution of (X, Y) is a bivariate lognormal distribution

$$f_{\mathrm{as}}(x, y) = \frac{1}{2\pi \ln^2 \sigma \sqrt{1-\rho^2}} \frac{1}{xy} \times \exp\left\{ -\frac{1}{2\ln^2 \sigma \sqrt{1-\rho^2}} (\ln^2 x/\mu - 2\rho \ln x/\mu \ln y/\mu + \ln^2 y/\mu) \right\}. \tag{23}$$

It should be noted that $\rho = \rho_{\ln X, \ln Y}$ is not the correlation of (X, Y) but of $(\ln(X), \ln(Y))$. The correlation of (X, Y) can be written down from (15) as

$$\rho_{X,Y} = \frac{G(1,1) - G(1)^2}{G(2) - G(1)^2} = \left(\frac{E(1,1)}{2 + 2K(2)} - \exp\{-(2 + 2K(2))t\} \right) \times (1 - \exp\{-(2 + 2K(2))t\})^{-1} \tag{24}$$

since $K(1) = -1/2$ for binary fragmentation. However, this has no simple general relation with ρ.

2.4. Conditional distributions

The conditional distributions (1) are from (23) and (9)

$$f_{\mathrm{as}}(y|X = x) = \frac{1}{\ln \sigma \sqrt{2\pi(1-\rho^2)}} \frac{1}{x} \exp\left\{ -\frac{1}{2(1-\rho^2)} \left(\frac{\ln(y\mu^{\rho-1}/x^\rho)}{\ln \sigma} \right)^2 \right\} \tag{25}$$

and the geometric expectation and geometric standard deviation conditioned on the size x of the adjacent fragment are $\mu_x = x^\rho/\mu^{\rho-1}$ and $\sigma_x = \sigma^{\sqrt{1-\rho^2}}$, respectively. The expectation conditioned on the adjacent fragment size x is

$$m_x = \frac{x^\rho}{\mu^{\rho-1}} \exp\left\{ \frac{1}{2} \ln^2 \sigma^{\sqrt{1-\rho^2}} \right\}. \tag{26}$$

2.5. The general case

Equation (10) generalized to n successive adjacent fragments,

$$\begin{aligned} \frac{\partial}{\partial t} N f_n(x_1, x_2, \ldots, x_n) &= N \int_{x_1}^{1} \mathrm{d}z\, \alpha(z) f_n(z, x_2, \ldots, x_n) B(z \to x_1) \\ &\quad + N \int_{x_n}^{1} \mathrm{d}z\, \alpha(z) f(x_1, x_2, \ldots, x_{n-1}, z) B(z \to x_n) \\ &\quad + N \sum_{i=1}^{n-1} \alpha(x_i + x_{n-i}) f_{n-1}(x_1, \ldots, x_{i-1}, x_i + x_{i+1}, x_{i+2}, \ldots, x_n) \\ &\quad - N \left(\sum_{i=1}^{n} \alpha(x_i) \right) f_n(x_1, x_2, \ldots, x_n) \end{aligned} \tag{27}$$

yields, in an n-dimensional analogue of transform (11), the equation

$$\begin{aligned} \frac{\partial}{\partial t} N G_n(p_1, p_2, \ldots, p_n) &= N G_n(p_1 p_2, \ldots, p_n)(K(p_1) + K(p_n) - n + 2) \\ &\quad + N \sum_{j=1}^{n-1} G_{n-1}(p_1, \ldots, p_{i-1}, p_i + p_{i+1}, p_{i+2}, \ldots, p_n) E(p_i + p_{i+1}). \end{aligned} \tag{28}$$

The solution of any G_n can be obtained iteratively by starting from $G_2 = G(p_1, p_2)$. However, the relevant solutions for the limit consideration are those with $p_2 = \cdots = p_{n-1} = 0$. These satisfy the recurrence relation

$$G_n(p_1, 0, \ldots, 0, p_n) = \frac{K(p_1) + K(p_n) + n - 1}{2K(p_1 + p_n) - K(p_1) - K(p_n) + n - 1} G_{n-1}(p_1, 0, \ldots, 0, p_n). \tag{29}$$

Applying (22) the correlations ρ_n for two fragments separated by $n - 2$ intermediate fragments are given by the recurrence relation $\rho_n = \rho_{n-1} - 1/t(n-1)$ or, when iterating from $\rho_2 = \rho$, (21),

$$\rho_n = 1 + \frac{\varepsilon}{2L_2 t} - \frac{1}{t}\sum_{j=1}^{n-1}\frac{1}{j}. \tag{30}$$

The correlations $\rho_n = \rho_{\ln X_1, \ln X_n}$ determine the limit distribution of $f_n(x_1, x_2, \ldots, x_n)$ which is an n-dimensional lognormal distribution for which the correlation matrix of the corresponding normal distribution can be written in terms of (30). The correlations ρ_{X_1, X_n}, $n > 2$, can be obtained from (24) with the recurrence relation

$$G_n(1, 0, \ldots, 0, 1) = \frac{n-2}{n + 2K(2)} G_{n-1}(1, 0, \ldots, 0, 1). \tag{31}$$

3. Discussion and conclusion

For the two simplest cases $B(x) = 1$ (random placement of the cut) and $B(x) = \delta(x - 1/2)$ (fragmentation into equal halves) the values of $\{\varepsilon, L_2, K(2), E(1,1)\}$ are $\{2 - \pi^2/6, 2, -2/3, 1/6\}$ and $\{\ln^2 2, \ln^2 2, -3/4, 1/4\}$, respectively. It is seen from (35) that the correlations ρ_n approach unity rapidly with increasing $t = \ln(N)$. On the other hand, as $K(2) > -1$ it is seen from (24) and (31) that for large t the correlations ρ_{X_1, X_n} approach the value

$$\frac{E(1,1)}{2 + 2K(2)} \prod_{j=3}^{n} \frac{j-2}{j + 2K(2)}. \tag{32}$$

Thus in the limit of large t the first values of $\rho_{X_1,X_2}, \rho_{X_1,X_3}, \rho_{X_1,X_4}, \ldots$ for $B(x) = 1$ and $B(x) = \delta(x - \frac{1}{2})$ are 1/4, 3/20, 9/80, 81/880, ... and 1/2, 1/3, 4/15, 8/35, ... respectively. For the case $B(x) = \delta(x - \frac{1}{2})$ the size of a fragment is $(\frac{1}{2})^k$, where k is the number of fragmentations. This gives by a variable change in $f(x, y)$ the distribution $f(k_1, k_2) =$ Prob (X has fragmented k_1 times and Y has fragmented k_2 times). This and the n-dimensional version are asymptotically multinormal distributions.

As the correlations ρ_n and ρ_{X_1, X_n} were obtained from the transforms G_n of the exact solutions of the equations (10) and (27) the results established for them are generally valid. No restrictions were imposed on $B(x)$ and non-vanishing correlations were found whenever $\alpha = 1$. Asymptotically vanishing correlations are possible for other choices of parameter $\alpha(x)$; this is at least the case for $\alpha(x) = x$ and $B(x) = 1$ which defines a spatial Poisson process restricted to $[0, 1]$. Any multidimensional initial distribution can be taken into account by multiplying G_n by the transform G_0 of the initial distribution; the lognormality of the limit distribution is not affected by the initial distribution. The generalization of (10) and (27) to describe a process where the number of progeny m is larger than 2 is formally straightforward by reformulating the equations in terms of adjacent m-tuples of fragments. The results suggest also new methods for the exact treatment of two- and three-dimensional fragmentation-generated cellular structures (Delannay and Le Caer 1994). The transition to spatial dimensions higher than one is non-trivial as fragments are not arranged into a sequence where each fragment has only one adjacent fragment on each side. The number of fragments adjacent to a given fragment will depend both on the geometry of the fragments and on probabilities $B(y \to x)$. This necessitates a tailored formulation of (27) or a method of ordering the fragments in the structure, that is, the specification of a mapping to the one-dimensional analogue.

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