Scaling Theory of Fragmentation

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We develop a scaling theory for linear fragmentation processes, for general breakup kernels characterized by a homogeneity index $\lambda > 0$. We discuss the existence of scaling, and show that the scaled cluster-size distribution $\phi(x)$ generally decays with the scaled mass x as $x^{-2}\exp(-ax^{\lambda})$, as $x \rightarrow \infty$. For small x, $\phi(x)$ approaches the log-normal form, $\exp(-a\ln^2 x)$, if the kernel has a small-size cutoff, and a power-law form in the absence of a cutoff. We also show that $\lambda < 0$ leads to a shattering transition. Finally, we outline the essential features of a nonlinear fragmentation process.

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Fragmentation is a relatively ubiquitous phenomenon that underlies processes such as polymer degradation,[1,2] breakup of liquid droplets,[3] and the crushing of rocks.[4,5] Recently, there has been renewed interest in fragmentation[6-8] which has paralleled recent progress in the complementary process of aggregation.[9-12] Much of the theoretical work is based on the description of fragmentation by a system of *linear* rate equations, i.e., it is assumed that the breakup process is driven by an external source. For certain classes of models, both scaling solutions and a number of exact solutions have been discovered.[6,7] Some of the exact solutions are quite complex, however, and a universal classification of the kinetics in terms of qualitative aspects of the microscopic process is still lacking.

Our goal, in this Letter, is to provide such a classification through a scaling formulation. We find that the asymptotic form of the cluster-size distribution at large size is strongly determined by the homogeneity index of the breakup kernel. In the small-size limit, we also obtain the general conditions on the kernel which gives rise to a log-normal cluster-size distribution, a form characteristic of a random multiplicative process, and which arises in many rock crushing processes.[13,14] We also discuss the criterion for the existence of a "shattering" transition, in which mass is lost to a phase of zero-size particles. Finally, we introduce a *nonlinear* fragmentation model, which is driven by repeated collisions between fragments. Scaling solutions of the rate equations are obtained, which reveal the differences between the linear and nonlinear processes.

In the rate-equation approximation, linear fragmentation is described by the integro-differential equation[6-8]

$$\frac{\partial}{\partial t}c(x,t) = -a(x)c(x,t) + \int_x^\infty c(y,t)a(y)f(x\,|\,y)dy. \quad (1)$$

Here $c(x,t)$ is the concentration of x-mers at time t, $a(x)$ is their overall rate of breakup, and $f(x\,|\,y)$ is the rate at which x is produced from the breakup of y. We consider homogeneous kernels for which $a(x)=x^{\lambda}$, thus defining the homogeneity index λ. Homogeneity also implies that $f(x\,|\,y)$ has the form $y^{-1}b(x\,|\,y)$. These restrictions on the kernels encompass virtually all cases of physical interest. Mass conservation imposes the condition $\int_0^1 xb(x)dx=1$, and the average number of fragments produced upon particle breakup, $\int_0^1 b(x)dx$, is taken to be finite.

To analyze Eq. (1), we write the familiar scaling *Ansatz* for the cluster-size distribution,[9-12]

$$c(x,t) \sim s^{-2}\phi(x/s), \quad (2)$$

where s is the typical cluster mass. The substitution of Eq. (2) into (1), and the definition of $\xi = x/s$, yield the scaling equations

$$\omega[2\phi(\xi)+\xi\phi'(\xi)] = -\xi^{\lambda}\phi(\xi) + \int_{\xi}^{\infty}\phi(\eta)\eta^{\lambda-1}b\left(\frac{\xi}{\eta}\right)d\eta, \quad (3)$$

$$\dot{s}s^{-(1+\lambda)} = -\omega, \quad (4)$$

where $\omega > 0$ is the separation constant, and the overdot denotes the time derivative. Equation (4) immediately yields $s \sim t^{-1/\lambda}$ for $\lambda > 0$. To find the asymptotic solution to Eq. (3), we convert it to a relation involving the moments of $\phi(\xi)$ by multiplying both sides by ξ^{α} and integrating over all ξ. Thus in terms of the moments of the scaling function, $m_{\alpha} = \int_0^\infty x^{\alpha}\phi(x)dx$, and the reduced breakup kernel, $L_{\alpha} = \int_0^1 x^{\alpha}b(x)dx$, we obtain

$$m_{\alpha+\lambda} = \omega\frac{1-\alpha}{L_{\alpha}-1}m_{\alpha}, \quad (5)$$

where explicit dependence on the kernel is contained only in L_{α}, and we are free to choose a normalization where $m_0 = m_1 = 1$.

Before analyzing this moment relation, we discuss the general conditions for which the scaling *Ansatz*, Eq. (2), is valid asymptotically. This justification rests on the linearity of the rate equations, which dictates that $c(x,t)$

must decay exponentially in time, or slower, for any value of x. This corresponds to moments which have a smooth, nonsingular, time dependence. On the other hand, for $\lambda < 0$, Eq. (4) predicts that there is a singularity in the time dependence of the moments at a finite time. This contradiction implies that the necessary condition for the existence of scaling is $\lambda > 0$.

The fact that $\lambda > 0$ is a sufficient condition for the existence of scaling is supported by all existing exact solutions and by simulation results, and we therefore assume this fact. However, it is possible to give a plausibility argument for the existence of scaling solutions for $\lambda > 0$. This argument is based on our first writing a "bare" moment relation directly from the rate equations, by defining $M_\alpha = \int_0^\infty x^\alpha c(x,t)dx$, and then converting Eq. (1) into the moment relation

$$\dot{M}_\alpha = (L_\alpha - 1) M_{\alpha+\lambda}. \tag{6}$$

On the other hand, from the scaling *Ansatz* of Eq. (2), the moments can be written as

$$M_\alpha \sim m_\alpha s^{-1+\alpha} \sim t^{(1-\alpha)/\lambda}, \tag{7}$$

which indeed is a consistent solution of Eq. (6). Furthermore, by using the fact $M_1 = 1$, one can show that Eq. (7) is the only asymptotic solution to Eq. (6), for a set of equidistant α values, $\alpha = 1 - k\lambda$. If we further assume "smoothness," wherein the form of M_α for arbitrary α interpolates smoothly between the moments defined on the discrete set, then the scaling solution substituted in Eq. (6) generally leads to (5), as $t \rightarrow \infty$.

To determine the behavior of $\phi(x)$, we now use Eq. (5) to compute the asymptotic form of the reduced moments for a set of equidistant α values, and then use the properties of the inverse Mellin transform to reconstruct the functional form of the scaling function. In this reconstruction, we assume that the leading behavior of m_α "corresponds" to the leading behavior of $\phi(x)$. For example, if for some $\phi(x)$ there exists a value α_c such that m_α is finite for $\alpha < \alpha_c$ and is infinite for $\alpha > \alpha_c$, then it is interpreted that $\phi(x)$ asymptotically behaves like the power law,

$$\phi(x) \propto x^{-1-\alpha_c}.$$

Although this is not mathematically precise, this correspondence is supported by available exact solutions, and it appears to be correct for most physically realizable fragmentation processes. In cases where such a correspondence does not apply, the comparison of the limiting forms of m_α can still be a valuable measure of the closeness of the corresponding $\phi(x)$'s. Therefore, in the following discussion of the asymptotic behavior of $\phi(x)$, it is understood either that this assumption is implied, or that moments which have the same leading behavior will correspond to $\phi(x)$'s that are "close."

For the large-x behavior of $\phi(x)$, we require m_α for large values of α. To obtain this latter quantity, we take $\alpha = k\lambda$, with k an integer, and iterate Eq. (5). Use of the fact that $m_0 = 1$ leads to

$$m_{k\lambda} = \omega^{k-1} \prod_{n=1}^{k-1} (n\lambda - 1) \left[\prod_{n=1}^{k-1} (1 - L_{n\lambda}) \right]^{-1}. \tag{8}$$

The asymptotic behavior of $m_{k\lambda}$ can now be obtained for a very large class of kernels which, for x near 1, i.e., the limit of production of large fragments, have the form

$$b(x) = b(1) + O((1-x)^\mu) \tag{9}$$

where $b(1) \geq 0$ and $\mu > 0$ are constants. Use of this in Eq. (8) and the employment of Stirling's approximation for the factorial function yields

$$m_\alpha \rightarrow c(\omega/e)^{\alpha/\lambda} \alpha^{[b(1)-1]/\lambda - 1/2} \alpha^{\alpha/\lambda}, \tag{10}$$

for $\alpha \rightarrow \infty$, where c is a constant.

This form for m_α is universal in the sense that the breakup kernel enters only in c, ω, and $b(1)$; this universality will enable us to deduce the general form of $\phi(x)$ in terms of the m_α's. To accomplish this, we consider first a simple exactly solvable "test" system for which the kernel satisfies Eq. (9). For this case, the exact connection between the moments and the scaling form of the cluster-size distribution can be found. The universal form of the moments then guarantees that this connection extends to arbitrary kernels which satisfy Eq. (9). The test system is one in which there is a uniform probability of fragment sizes in any breakup event, corresponding to $b(x) = 2$. The exact solution for the cluster-size distribution[2] can be written in the following scaling form, in the long-time limit:

$$\phi(x)^{\text{exact}} \sim \frac{2\mathcal{M}}{\Gamma(1+2/\lambda)} \exp(-x^\lambda), \tag{11}$$

where $\mathcal{M}$ is the mean mass in the initial condition. The corresponding moments are

$$m_\alpha^{\text{exact}} \sim \frac{2\mathcal{M}}{\lambda\Gamma(1+1/\lambda)} \Gamma\left(\frac{\alpha+1}{\lambda}\right) \rightarrow \text{const} \times \left(\frac{1}{\lambda e}\right)^{\alpha/\lambda} \alpha^{1/\lambda - 1/2} \alpha^{\alpha/\lambda}. \tag{12}$$

To compare Eq. (10) with (12), we first rewrite Eq. (10) in terms of the shifted variable, $\alpha' = \alpha + b(1) - 2$,

$$m_\alpha \rightarrow c'(\omega/e)^{\alpha'/\lambda} (\alpha')^{1/\lambda - 1/2} (\alpha')^{\alpha'/\lambda} = c''(\omega\lambda)^{[\alpha+b(1)-2]/\lambda} m_{\alpha+b(1)-2}^{\text{exact}}, \tag{10'}$$

where the second half follows by direct comparison with Eq. (12). By using the Mellin transform property that if the moments m_α are obtained from $\phi(x)$, then the moments $a^{-(\alpha+\sigma+1)}m_{\alpha+\sigma}$ are obtained from $x^\sigma\phi(ax)$, we find that $\phi(x)$ has the asymptotic form

$$\phi(x) \sim x^{b(1)-2}\exp(-ax^\lambda), \quad x \to \infty. \tag{13}$$

The generic case $b(1)=0$ includes kernels with power-law decays, exponential decays, and finite cutoffs for x near 1.

This result for $\phi(x)$ can be extended to general homogeneous kernels by our noticing that the controlling factor in Eq. (10), $\alpha^{\alpha/\lambda}$, is responsible for the controlling factor, $\exp(-ax^\lambda)$, in $\phi(x)$. This feature is *universal* for any homogeneous kernel, regardless of whether or not condition (9) is satisfied.

For small x there is a lesser degree of universality, as might be anticipated, since the small-mass tail is not influenced by clusters of the typical size. We do find, however, that there are two possible generic scaling forms for $\phi(x)$ at small x, whose applicability depends on whether or not the moments $L_{-\alpha}$ exist for all α. Consider first kernels that are cut off at small fragment sizes, i.e., $b(x)$ decays faster than $\exp(-x^{-\mu})$ for some $\mu > 0$ as $x \to 0$, so that $L_{-\alpha}$ is finite for all α. To study the small-x limit of $\phi(x)$, we now choose $\alpha = 1-k\lambda$ in Eq. (5), and iterate it to arrive at the analog of Eq. (8), namely,

$$m_{1-k\lambda} = \omega^{-k}\prod_{n=1}^{k}(L_{1-n\lambda}-1)\left[\prod_{n=1}^{k} n\lambda\right]^{-1}. \tag{14}$$

As an illustrative example, consider a sharp cutoff on the kernel, i.e., $b(x)$ vanishes for $x < x_0$, with $0 < x_0 < 1$. Then $L_{-\alpha}$ has the controlling factor $x_0^{-\alpha}$ for large α, and the corresponding controlling factor of $m_{-\alpha}$ is

$$\exp\left[\frac{\ln x_0^{-1}}{2\lambda}\alpha^2\right], \tag{15}$$

which is the Mellin transform of the log-normal distribution. Thus by taking the inverse Mellin transform, we predict that the controlling factor of $\phi(x)$ is

$$\phi(x) \sim \exp\left[-\frac{\lambda}{2\ln x_0}(\ln^2 x)\right], \quad x \to 0. \tag{16}$$

More generally, we can show that for an arbitrary kernel, the controlling factor in $m_{-\alpha}$ will generally diverge at least as fast as $\exp(c\alpha^2)$, and this implies that $\phi(x)$ will decay as $\exp[-(\ln^2 x)/4c]$, or slower, as $x \to 0$.

Let us now consider kernels for which there is no small-size cutoff, in a single fragmentation event. This is typified by kernels with a power-law decay for small x, i.e., $b(x) \sim x^\nu$. In this case, it follows from Eq. (14) that m_α diverges whenever L_α diverges, and this occurs for α less than a critical value α_c, which is less than 0, since m_0 is finite. For α close to α_c we keep only the leading term in Eq. (5) to give

$$m_\alpha = L_\alpha \frac{m_{\alpha_c+\lambda}}{\omega(1-\alpha_c)} \sim \text{const} \times \int x^\alpha b(x)dx. \tag{17}$$

This implies that $\phi(x)$ coincides with $b(x)$, so that

$$\phi(x) \sim x^\nu, \quad x \to 0. \tag{18}$$

Because clusters of indefinitely decreasing size are produced in fragmentation, there is the possibility of a shattering transition, in which mass is "lost" to a phase of zero-mass particles. This is reminiscent of gelation, where the mass of finite-size clusters is lost to a phase consisting of an infinite gel molecule. Both gelation and shattering are signaled by the condition $\dot{M}_1 < 0$. As first discussed by McGrady and Ziff[6] in the context of a particular fragmentation model, shattering was found to occur when $\lambda < 0$, and in this case, scaling breaks down. We now show generally that $\lambda < 0$ is a necessary and sufficient condition for the existence of shattering.

To locate the shattering transition, we examine Eq. (6) in the limit $\alpha \to 1$. Since L_α approaches 1 from below as $\alpha \to 1^+$, while $M_\alpha > 0$, $\dot{M}_1 \le 0$ in general. In the limit $\alpha \to 1$, the existence of shattering depends only on having $M_{1+\lambda+\epsilon}$ diverge faster than $1/\epsilon$ as $\epsilon \to 0$. By definition, M_α is nondecreasing as α decreases, at fixed time. This fact, together with $M_1 < \infty$, implies that M_α can diverge only for $\alpha < 1$. Thus a necessary condition for shattering is $\lambda < 0$. It is also a sufficient condition because if the converse were true, then from Eq. (6), $M_{1+|\lambda|}$ would go to zero at a finite time, which contradicts the fact that $c(x,t)$ cannot vanish at a finite time.

To summarize, scaling provides a comprehensive description for the cluster-size distribution of linear fragmentation processes. For large x, $\phi(x)$ has the universal form $\exp(-ax^\lambda)$, where $\lambda > 0$ is the homogeneity index of the overall breakup rate, while for small x, $\phi(x)$ may either have a log-normal tail, for kernels with small fragment cutoff, or a power-law decay for kernels with no cutoff. We now wish to extend this scaling description to a new fragmentation model, in which repeated collisions between clusters, as might be envisioned in crushing, are the source for fragmentation.[15] This collision-driven process is inherently nonlinear, as the collision rate varies as the square of the particle concentration (at low concentrations). We map out basic features of this model, and determine essential differences with linear fragmentation.

The rate equations are

$$\frac{\partial}{\partial t}c(x,t) = -c(x,t)\int_0^\infty K(x,y)c(y,t)dy + \int_0^\infty dz\int_x^\infty K(y,z)B(x\,|\,y,z)c(y,t)c(z,t)dy. \tag{19}$$

The first term accounts for the loss of x because of collisions of x-mers with the remaining particles in the system, at a rate specified by the homogeneous collision kernel, $K(ax,ay)=a^{\lambda}K(x,y)$. The second term accounts for collisions in which a y-mer and a z-mer produce x-mers at a rate specified by the breakup kernel, $B(x\,|\,y,z)$. Mass conservation imposes the condition $y=\int_0^y x \times B(x\,|\,y,z)dy$, and also $B(ax\,|\,ay,az)=a^{\alpha}B(x\,|\,y,z)$, with $\alpha=-1$, if the kernel is homogeneous.

With use of the scaling *Ansatz*, Eq. (2), in Eq. (19), the typical size s obeys $\dot{s}s^{-\lambda}=-\omega$. Thus s vanishes in a finite time for $\lambda<1$, in contrast to linear fragmentation, where s vanishes in a finite time only for $\lambda<0$. To determine the cluster-size distribution, we make the simplification wherein the particle undergoing breaking always splits into two equal halves, so that the mass x can be written as $x=2^{-n}$, with n an integer. This equal-splitting feature still retains the essential nonlinearity of the collision-induced process, while being simple enough to be tractable. By numerical integration of the rate equations, together with the use of consistency arguments, it appears that the scaling holds for $\lambda>\lambda_c$, with $\lambda_c=0$ if both particles break upon collision, $-1<\lambda_c<0$ if only the larger particle splits, and $\lambda_c=1$ if only the smaller particle splits. Generally, the scaling regime coincides with the range of λ for which there is no shattering transition.

In the scaling regime, the asymptotic solutions to the rate equations are,

$$c_n(t)\sim\begin{cases}2^n\exp[-2^{-n\lambda}t^{\lambda/(\lambda-1)}], & \text{larger splits,}\\ 2^{\lambda n}/t, & \text{smaller splits.}\end{cases}$$

By matching to the scaling form of the cluster-size distribution we then obtain

$$\phi(x)\sim\begin{cases}\exp(-x^{\lambda/2})/x^2, & \text{both split,}\\ \exp(-x^{\lambda})/x^2, & \text{larger splits,}\\ x^{-(1+\lambda)}, & \text{smaller splits,}\end{cases}$$

as $x\to\infty$. Thus in contrast to linear fragmentation, the cluster-size distribution at large masses can have a power-law tail. This feature should be useful in differentiating whether linear or nonlinear mechanisms underlie a particular fragmentation process.

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