

10. Fragmentation

Sidney Redner[†]

10.1 Introduction

The breaking up of particulate material into smaller fragments is a ubiquitous process that underlies many natural phenomena and technological processes. At the geological time scale, fragmentation is responsible for the distribution of fragment sizes that appear on beaches, on mountain boulder fields, and in soils.^[1-4] Related degradation processes are responsible for the size distribution of lunar geological features^[5-7] and the size distributions of asteroids.^[8-10] More practical examples of fragmentation occur in mineral processing,^[11-15] where the size reduction of raw ore is the initial step that must be performed in order to extract the valuable minerals. This breakage process is energy intensive and generally quite inefficient. Owing to the non-trivial fraction of all energy use that goes into mineral processing, considerable effort has been devoted to the optimal design of ore processing machinery.

In addition to the obvious realizations of fragmentation in rock fracture, there are many other situations where various forms of size reduction and fragmentation occur. At a molecular level, the breaking of individual chemical bonds underlies polymer degradation phenomena.^[16-22] In combustion and in other types of dissolution phenomena, as the surface of the consumed object recedes, irregularities form which ultimately lead to the breaking up of the material.^[23-27] This process is responsible for the size distribution of the residual ash, and an understanding of its properties is important in pollution control. The breakup of liquid droplets by agitation^[28-31] is an example where surface tension needs to be surmounted to cause fragmentation, and the resulting aerosol formation has a myriad of practical applications. Within the context of fluids, the breakup of eddies and vortices in turbulent fluid flows may also be viewed as fragmentation processes,^[32] where the entity undergoing breakup is the flow field itself. Thus in a variety of forms, fragmentation is the primary mechanism for many dynamic phenomena.

[†] Center for Polymer Studies and Department of Physics, Boston University, Boston, MA 02215, USA.

In most realizations of fragmentation one is unable to observe a given breakup event in detail, but rather, one observes the *distribution* of fragment sizes that results from the breakup process. Theoretical treatments of fragmentation have therefore been focused primarily on either predicting the fragment size distribution, based on knowing (or assuming) certain facts about the instantaneous breakup process, or conversely, inferring details about the breakup process based on experimental observations of the fragment size distribution. There are a wide variety of possible instantaneous breaking mechanisms which depend on the material properties and geometry of the object undergoing breaking, and also on the energy input to the system. These details determine the nature of the fragment size distribution in an instantaneous breakup event. For a continuously evolving fragmentation process, however, particle breakup eventually takes place on all size scales. This feature suggests the application of statistical approaches in which the fragment size distribution from a single breakup event serves to define basic parameters in the dynamical rate equations that govern continuous fragmentation. The goal of this chapter is to investigate the nature of the fragment size distributions that arise in continuous fragmentation processes through the solution to these rate equations.

There already exists a vast literature on fracture^[33-50] and fragmentation^[51-66] and a wide range of phenomena have been investigated, both theoretically and experimentally. Interestingly, what are regarded as immutable and basic facts in the literature of one subfield are unknown or apparently not appreciated in other subsets of the literature. I will therefore attempt to give a coherent presentation of some of the basic results which are representative of the broad spectrum of physical realizations of fracture and fragmentation. In section 10.2, I first review some basic facts about instantaneous fracture to illustrate the types of fragment size distributions that can occur and to discuss the basic parameters that influence the form of the distribution. The available data suggests that for a wide variety of materials and fracture mechanisms, there is a power-law dependence of the relative concentration of fragments on their size over a non-negligible range.^[33,40-47] It is intriguing that the exponent of this power law falls within a relatively narrow range of values for a wide variety of systems. This distribution of fragment sizes in instantaneous breakup serves as a basic ingredient in determining the dynamics of continuous fragmentation. Thus it is important to use the experimental observations to develop mathematical models that have a realistic basis.

In section 10.3, I will elucidate some of the recent progress made in understanding the nature of the fragment size distributions that occur in continuous fragmentation. I will focus primarily on the investigation of this problem by the rate equations.^[51,55,58-66] The rate equations are an approximation of a mean-field character, as fluctuations in the system are ignored. That is, fragments are assumed to be distributed homogeneously at all times throughout the system, as might actually occur in a milling process, and the variability of cluster shapes is ignored. Thus the mass is the only dynamical variable that characterizes a fragment in

the rate equations. Although there are limitations inherent in this approach, the rate equations do provide a comprehensive theoretical description for fragmentation which do compare well with certain experimental results. Moreover, the rate equations may serve as the starting point for further theoretical refinements.

I will first outline some of the primary features that emerge from the exact solutions of the rate equations for a particularly simple class of physically-motivated models.^[58-60] I will then emphasize recent work in which scaling analysis is applied to extract asymptotic solutions to the rate equations.^[63,64] This is a relatively simple, yet powerful method which provides a universal classification scheme for the rate equation solutions. The applicability of scaling rests on the observation that the fragment size distribution is often characterized by a single typical cluster size, which is a decreasing function of time. Consequently, it is reasonable to expect that the fragment size distribution will be a function *only* of the ratio of the size to the typical size. From this physical expectation, it is possible to determine the possible asymptotic forms for the cluster size distribution quite simply. I also discuss a special regime of behaviour, in which small clusters are more likely to break up than large clusters, that leads to "shattering".^[51,60,63,64] This is a mathematical pathology which describes the interesting situation where mass is lost to a shattered phase consisting of an infinite number of zero mass particles. The general features of the universality and scaling of the fragment size distribution and the shattering phenomenon have parallels in the inverse process of aggregation.^[67-76]

In section 10.4, I give a brief summary and discuss some possible future research directions where the current theoretical understanding is incomplete. For example, the role of spatial inhomogeneities is largely unexplored, but clearly is a central aspect in geological fragmentation processes. In addition, while the widely-accepted assumption of the linearity of continuous fragmentation processes appears to be appropriate for certain situations, linearity is clearly inappropriate in cases where fragmentation is driven primarily by the particles in the medium rather than by an external source. The development of models to treat the non-linear aspects of fragmentation may be a rewarding new area of investigation.

10.1.1 Instantaneous fracture

Upon the sudden introduction of a sufficient strong external energy source, an object can break into smaller fragments, whose distribution is the primary observational evidence for the nature of the breakup process. For brittle materials, breaking occurs when the applied stress exceeds the fracture stress, which is often close to the point where linear response breaks down. According to the classical Griffith theory,^[77] the magnitude of this fracture stress depends on the distribution of pre-existing flaws, or cracks, in the material. Owing to the concentration of stress at a crack tip, the fracture stress of a flawed material is typically much smaller than the stress needed to break atomic bonds. For example, for a two-dimensional object containing a single linear crack of length L , the fracture stress σ is given by $\sigma = (2\gamma E/L)^{1/2}$, where γ is the surface free energy per unit crack surface area, and

E is Young's modulus of the material. Thus in a finite volume of brittle material, the largest crack in the object determines its fracture stress. Since the energy cost of a crack is proportional to an area, while the strain energy is proportional to a volume, the amount of energy available for crack formation at a fixed stress decreases as the particle size decreases.^[11,78] Thus under conditions of fixed energy density, one has the general fact that large objects are more susceptible to breaking than small objects. This monotonic size dependence of the overall breakup rate of an isolated object is one of the basic ingredients in the theoretical treatment of continuous fragmentation.

Interestingly, in the crushing of rocks, the energy is typically furnished in the form of uniaxial compression, or perhaps in the form of shear for the case of comminution. However, under a localized compression, there is a redistribution of stresses so that a considerable portion of an object of finite-size is under tension in a direction perpendicular to the external compression.^[79] This tensile stress can open cracks further, leading to ultimate failure. This mechanism appears to be the normal mode of breaking in materials such as rocks.

There is considerable well-documented data for the distribution of fragment sizes in a variety of realizations of instantaneous fracture, ranging from the over-compression of glass spheres,^[41-43] to high-velocity impacts of small projectiles onto fixed rock targets.^[46,47] On the basis of these experiments, the possible outcomes for the fracture event are often classified into three broad regimes of behaviour depending on the energy input and on the geometrical features of the experiment. While the terminology is not universal, the three regimes may be written as:^[11]

- abrasion (or surface erosion)
- cleavage (transition behaviour)
- destructive breaking (or shattering).

The corresponding particle size distributions for each of these regimes are sketched in fig. 10.1. In abrasion, the breaking mechanism is applied specifically to the surface, or the energy intensity is sufficiently small that only a small flecks are removed from the surface of the object. This leads to a bimodal fragment distribution, consisting of coarse product and fine product components. In cleavage, the energy input is just sufficient to propagate of the order of one crack through the sample. Typically, then, the resulting fragments are of the same order in size as the original object. Finally, in destructive breaking, enough energy has been imparted to the system so that many regions of the material are stressed beyond the breaking point. This leads to a wide range of fragment sizes being produced, which are typically all much smaller than the size of the original object. While this classification is a useful scheme to broadly characterize instantaneous breaking, it is highly idealized. In real breaking processes, there may be a continuum of outcomes as a function of the energy input rather than a sharply defined transition between two well-defined regimes.

For treating continuous fragmentation, the size distribution that arises from instantaneous breaking is a fundamental building block upon which theories for

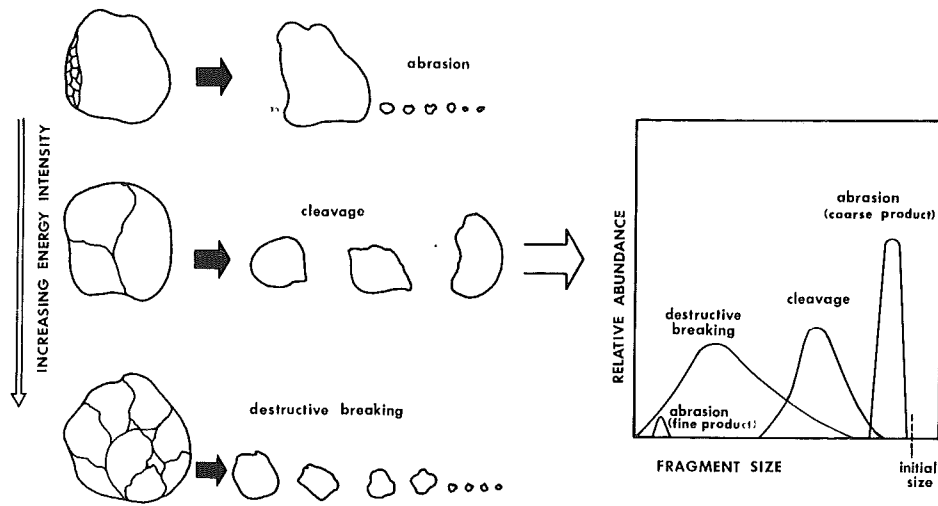


Figure 10.1 Schematic representations of the different mechanisms for instantaneous particle fracture and sketches of the corresponding fragment size distributions (adapted from ref. 11).

Continuous breakup can be constructed. There are a number of equivalent forms for expressing the size distribution, and the alternatives employed depend, to a large degree, on the nature of the experiment and/or on convention. Experimentally, it is often most convenient to consider $M(r)$, the cumulative mass of fragments or linear dimension less than r . This quantity is obtained naturally in sieving measurements. Alternatively, this type of measurement also provides $N(x)$, the cumulative number of fragments with mass greater than x . Finally, many theoretical treatments often express the size distribution in terms of $c(x) dx$, the concentration of fragments with mass between x and $x + dx$. In the limit of small fragment masses (or radii) and for a wide variety of physical systems, these distributions appear to be well-described by the power-law forms,^[33]

$$\begin{aligned} M(r) &\propto r^\alpha, \\ N(x) &\propto x^{-(1-\alpha/3)}, \\ f(x) dx &\propto x^{-(2-\alpha/3)}, \end{aligned} \quad (10.1)$$

where the exponents characterizing the latter two distributions follow by a simple change of variables from the distribution for $M(r)$. In many experiments on instantaneous and highly energetic breaking, exponent values between 0.5 and 1.0 have been quoted for α .^[41-47,55] For certain cases, this power law extends over two decades of fragment sizes, in the small size limit. This power-law behaviour appears to be a universal aspect of instantaneous and destructive breaking of brittle

objects.

Many of the theories that have been developed to describe the fragment size distribution arising in instantaneous breaking are probabilistic in nature. They are based on the idea that pre-existing flaws are distributed throughout a brittle material. When the object is stressed far beyond its fracture point, these flaws are activated and propagate until they reach the boundary of the material or until another crack is encountered. This ultimately leads to the breaking off of fragments whose size distribution is controlled by the location and geometry of the flaws. Perhaps the most detailed investigation in this spirit is that of Gilvarry.^[41] Under the assumption of a purely random distribution of flaws, the mass fraction of fragments with linear size less than r was found to vary as

$$M(r) = 1 - \exp(-r/k_1 - r^2/k_2 - r^3/k_3), \quad (10.2a)$$

where the coefficients k_1 , k_2 , and k_3 are proportional to the densities of linear, area, and volume flaws, respectively. In the case where only one class of flaws is predominant, the above distribution reduces to the exponential form

$$M(r) = 1 - \exp(-r^\beta/K). \quad (10.2b)$$

This is also termed the Weibull^[37] or Rosin-Rammler^[39] distribution, with the exponent β the index of the distribution. Refinements of the Gilvarry approach have been proposed by various authors in which the probabilistic aspects of the original Gilvarry theory are treated more carefully. Klimpel and Austin^[45] thus derived the following form for $M(r)$

$$M(r) = 1 - \left(1 - \frac{r}{K}\right)^{n_1} \left(1 - \left(\frac{r}{K}\right)^2\right)^{n_2} \left(1 - \left(\frac{r}{K}\right)^3\right)^{n_3}, \quad (10.2c)$$

which gave excellent agreement with experiments over nearly the entire size range. Here the parameters n_i 's are constants which depend on the densities of edge, area, and volume flaws, respectively.

In the small size limit, these distributions all reduce to the power law

$$M(r) \propto r^\gamma, \quad (10.2d)$$

with the exponent $\gamma = 1$. (This apparently universal form is often called the Gaudin-Schuhmann distribution in the mining engineering literature.^[40,44]) Considerable data for the instantaneous fracture of an isolated object are in good agreement with the hypothesis that $M(r) \sim r^1$ (fig. 10.2). Although this power law is found to hold over a substantial range, the upper limit of linearity for the cumulative mass is typically 0.1, or less. The full Gilvarry, or Klimpel-Austin forms provide a better fit for this remaining mass fraction which belongs to the largest fragments. It is worth noting that in the Gilvarry experiments, these largest fragments are contiguous to the surface of the initial object. Gilvarry hypothesized

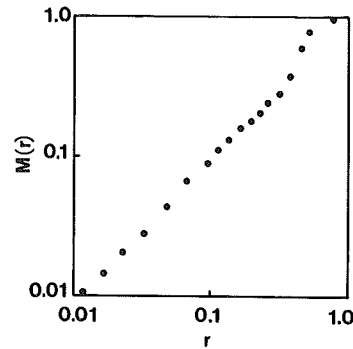


Figure 10.2 Fragment size distribution for 2.54 cm diameter glass spheres subjected to a compressive load far beyond the fracture stress. Shown is the cumulative fraction of mass, $M(r)$, whose linear dimension is smaller than r (from ref. [43]). The slope of the linear portion of the data is unity.

that these “surface” fragments may have a different distribution from that of the smaller, interior fragments. The possibility that surface, or finite size effects play a role in the fragment size distribution is appealing, but this aspect has not been seriously investigated.

The experimental facts outlined above serve as a basic framework from which one can develop theories for continuous fragmentation.

10.2 Continuous fragmentation

10.2.1 The rate equations

Having outlined some basic facts about instantaneous fracture, we now build on these results to investigate the basic features and the results which follow from the rate equation approach to continuous fragmentation. Implicit in this viewpoint is the assumption that the form of the fragment size distribution that arises in a single isolated breakup event continues to hold in continuously-evolving fragmentation. For this dynamic process, we now denote by $c(x, t)$ the concentration of clusters of mass x at time t . Notice that the concentration is taken to be spatially uniform, an assumption which might be realized in a strongly mixing milling process. Furthermore, by keeping only the cluster mass as the basic variable, one is ignoring potential effects induced by the variability in fragment shapes.

According to these conditions, the evolution of $c(x, t)$ may be described by the linear integro-differential equation^[58-64]

$$\frac{\partial c(x, t)}{\partial t} = -a(x) c(x, t) + \int_x^\infty c(y, t) a(y) f(x|y) dy. \quad (10.3a)$$

Here $a(x)$ is the *overall* rate at which x breaks, i.e., $a(x) dt$ is proportional to the probability that a cluster of mass x breaks in a time interval dt , while $f(x|y)$ is the *relative* breakup rate, i.e., the conditional probability at which x is produced from the breakup of y . The first term on the right hand side of eq. (10.3a) therefore accounts for the loss of clusters of mass x due to their breakup, while the second term accounts for the gain of x -clusters by the breakup of particles with mass larger than x .

In the case of purely binary breakup, where each breakup event produces exactly two fragments, the rate equations may be written in the alternative and convenient form,

$$\frac{\partial c(x, t)}{\partial t} = -c(x, t) \int_0^x F(y, x-y) dy + 2 \int_x^\infty c(y, t) F(x, y-x) dy. \quad (10.3b)$$

Here $F(x, y)$ is the rate at which a cluster of mass $x + y$ breaks up into two fragments, one of mass x and the other of mass y . The breakup rates in eqs. (10.3) are related by $a(x) = \int_0^x F(y, x-y) dy$, and for the case of binary breakup, $f(x|y) = f(y-x|y)$.

The cluster size distribution that results from eq. (10.3) is determined by the details of the kernels $a(x)$ and $f(x|y)$ (or, equivalently, by $F(x, y)$). With their form left unspecified, it is only possible to find the general conditions for which the fragment size distribution approaches a limiting form at long times. Consequently, there has been considerable work on solving the rate equations for specific, physically-motivated breakup kernels. Exact solutions have been found for models which are meant to account for situations such as ore comminution, powder crushing, and fly ash formation. In particular, Ziff and coworkers have recently solved the rate equations of fragmentation for a relatively wide class of models.^[58-60] Their work is notable for the general insights gained which parallel recent progress made in the investigation of aggregation phenomena. An illustrative example of this type of solution will be given in the next subsection.

By focusing on specific examples, however, it becomes difficult to appreciate the range of possible behaviours. I will therefore outline a scaling approach to fragmentation phenomena, in which universal features of the fragment size distribution emerge clearly.^[63,64] In order to apply scaling, I will henceforth specialize to the case of systems with breakup rates that are homogeneous functions of the fragment size. Homogeneity implies that the relative size, rather than the absolute size is the basic parameter which characterizes a breakup event. Therefore the overall breakup rate depends on the mass of a fragment as $a(x) = x^\lambda$, where λ is termed the homogeneity index. According to the discussion of the last section, we expect that λ is positive in general. Homogeneity also implies that $f(x|y)$ depends only on the ratio of the mass of the product to the mass of the initial cluster, i.e., $f(x|y) \propto y^{-1} b(x/y)$. From the experimental data shown previously, $b(x)$ is often described by a power-law form in the small- x limit. By definition, the integral

$\int_0^\infty b(x) dx$ equals the average number of fragments produced in a single breakup event, and mass conservation imposes the condition $\int_0^1 x b(x) dx = 1$.

Ostensibly, the restriction to homogeneity is not very restrictive, as a wide range of physical systems appear to be described by homogeneous kernels. Possible limitations associated with the restriction to homogeneity which will be discussed in section 10.4. In the discussion that follows, we will outline both exact and scaling solutions to fragmenting systems with homogeneous breakup rates.

10.2.2 Illustrative exact solutions

Since the rate equations for continuous fragmentation are linear, they are soluble, in principle, for arbitrary breakup kernels. Ziff and McGrady have outlined a formal procedure which yields the general solution to the rate equations for the case of binary breakup.^[58,59] A salient feature of these solutions is that for fixed x , the concentration $c(x, t)$ decays purely exponentially in time as $t \rightarrow \infty$. This fact can be seen directly from the rate equations. At sufficiently long times, the gain term in the rate equations can be neglected for any finite value of x . Consequently, the leading behaviour for $c(x, t \rightarrow \infty)$ is proportional to $e^{-a(x)t}$. The role of the gain term is to introduce power-law modifications to this asymptotic exponential behaviour. Building on this insight, one way to obtain the solution for all times is to write the full solution as a product of the long-time solution times a power series in t , in which each coefficient is a function of x . This leads to recursion relations for the series coefficients which can be solved in closed form for various cases.

To illustrate the important features of the fragment size distribution, let us consider one very simple class of models in which there is binary breakup, with the breakup rate depending only on the size of the object breaking up, and not on the sizes of the product fragments.^[58] Using the description given in eq. (10.3b), this corresponds to $F(x, y)$ depending only on the combination $x+y$. For a homogeneous system, one therefore has $F(x, y) \propto (x+y)^{\lambda-1}$. Models of this kind have been invoked to describe the breakup of polymer chains under the action of shear or by chemical degradation, for the case where $\lambda = 1$. This describes the situation where each bond in the chain breaks at a constant rate. For general values of λ , the solutions to the rate equations illustrate how the overall breakup rate manifests itself in the form of the cluster size distribution.

For $F(x, y) = (x+y)^{\lambda-1}$, eq. (10.3b) becomes

$$\frac{\partial c(x, t)}{\partial t} = -x^\lambda c(x, t) + 2 \int_x^\infty y^{\lambda-1} c(y, t) dy, \quad (10.4)$$

or by differentiating with respect to x ,

$$c_{xt}(x, t) = -x^\lambda c_x(x, t) - (\lambda + 2)x^{\lambda-1} c(x, t), \quad (10.5)$$

where the subscripts denote partial differentiation. In view of the limiting asymptotic exponential form for $c(x, t)$, we attempt a solution of the form

$$c(x, t) = A(t) e^{-B(t)x^\lambda}. \quad (10.6)$$

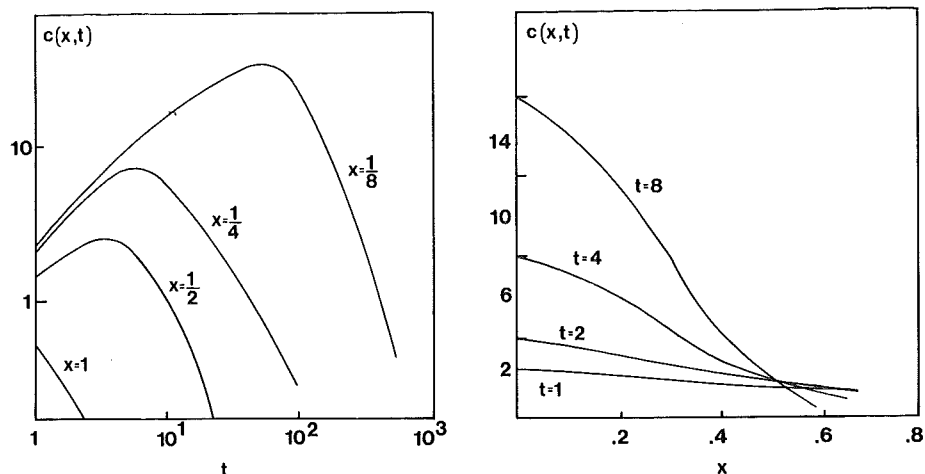


Figure 10.3 Sketch of the fragment size distribution, $c(x,t) = 2te^{-tx^2}$, based on the exact solution to the rate equations for the case of binary breakup with $F(x,y) = (x+y)$ (from ref. [58]). Shown is, (a) the distribution as a function of time for various values of mass x , and (b) the distribution as a function of x at various times.

This yields soluble ordinary differential equations for $A(t)$ and $B(t)$, from which one obtains a special solution for $c(x,t)$,

$$c(x,t) = (1 + t/s)^{2/\lambda} e^{-(s+t)x^\lambda}, \quad (10.7)$$

where $s = B(0)$. By the linearity of the rate equations, the general solution for $c(x,t)$ can be written as a linear combination, that is, an integral transform of eq. (10.7), which, in turn, can be expressed in terms of special functions.

To gain some intuition for the fragment size distribution, consider $c(x,t)$ in the long time limit. This gives the asymptotic behaviour,

$$c(x,t) \propto t^{2/\lambda} \exp(-tx^\lambda). \quad (10.8)$$

A sketch of this fragment size distribution is shown in fig. 10.3, both as a function of time for fixed size, and as a function of size at fixed times. Qualitatively, the behaviour is in accord with simple-minded intuition. The population of a given (small) size, x , initially grows due to the breakup of larger clusters. Eventually, however, this size population decays when the production of x diminishes due to the depletion of larger clusters. This decay is asymptotically exponential, with the decay time varying inversely in the particle size. For the distribution at fixed time, there is a steepening near the origin as a function of time, reflecting the eventual predominance by very small clusters. This small mass tail often has a power-law

form, as discussed previously. Notice also that this asymptotic form for $c(x, t)$ becomes pathological as $\lambda \rightarrow 0$. This is a signal of the shattering transition, which will be discussed in a later section.

10.2.3 Scaling formulation

As discussed in the introduction, the scaling approach is a simple but very powerful conceptual tool to analyze the solutions to the rate equations. According to scaling, the ratio of the size to the typical size is the *only* parameter which characterizes a fragmenting system in the scaling limit. This scaling behaviour is already evident from the exact solutions treated above. There are a number of important reasons to emphasize scaling solutions. First, scaling generally provides the simplest route to the asymptotic solution of the rate equations, especially in complicated situations, where exact solutions require considerable technical expertise. This simplification follows because the scaling ansatz reduces a two-variable problem to a single variable problem. This reduction is an important motivating factor for using the scaling approach for dynamic phenomena in general. Second, a scaling solution is universal in that it is independent of initial conditions. Thus scaling provides a universal classification of the solutions to the rate equations for an encompassingly-wide wide class of fragmenting systems.

The scaling ansatz for the cluster size distribution can be written as^[67,73-76]

$$c(x, t) = s^{-2} \phi(x/s(t)), \quad (10.9)$$

where $s(t)$ is the typical (time-dependent) cluster mass, and the exponent -2 is required by mass conservation. Thus the cluster concentration at long times depends on the ratio of the mass to the typical mass, rather than being a function of mass and time separately. A scaling ansatz of this general spirit forms the basis for the asymptotic analysis for a wide variety of dynamic phenomena, and has had extensive success in processes such as aggregation.

In our scaling approach, it is easiest to first compute the *moments* of the fragment size distribution, and then reconstruct the distribution, rather than computing the distribution directly.^[63,64] For this purpose, we define the α th moment of the cluster size distribution, $M_\alpha(t)$, and the α th moment of the scaling function, m_α , by

$$M_\alpha(t) = \int_0^\infty x^\alpha c(x, t) dx, \quad m_\alpha = \int_0^\infty x^\alpha \phi(x) dx. \quad (10.10)$$

These are just the Mellin transforms^[80] of $c(x, t)$ and $\phi(x)$, respectively (except for a trivial shift of unity in the definition of α). From these definitions, $M_0(t)$ is the total number of clusters, and $M_1(t)$ is the total mass in the system at time t . In these definitions, there are two free parameters which arise from the amplitudes of $s(t)$ and ϕ in the scaling ansatz. Without loss of generality, we choose $m_0 = m_1 = 1$ in order to fix these two free parameters.

For systems which obey scaling, the “bare” and “scaled” moments are related by

$$M_\alpha(t) = m_\alpha s(t)^{\alpha-1}. \quad (10.11)$$

Thus all the moments of the cluster size distribution are accounted for by a single size scale $s(t)$, and the special case $\alpha = 0$ yields,

$$s(t) = m_0/M_0(t). \quad (10.12)$$

Thus for a system with a unique typical size scale, the average size is inversely proportional to the average number of clusters. The relation between the scaled moments m_α and the bare moments $M_\alpha(t)$ follows solely from the ansatz (10.9), therefore this relation is common to fragmentation, aggregation, and other dynamic phenomena which are described by scaling.

A crucial step in our scaling analysis is to relate the moments of the cluster size distribution to the corresponding distribution itself. This relation is embodied by the following correspondence. First, by scaling, we compute the moments m_α directly for a discrete set of equidistant α values. We then invoke “smoothness”, in which we assume that the form of the moments defined on the discrete set $\{\alpha\}$ continues to hold for all real values of α . Finally, the functional form of the scaling function for the cluster size distribution, $\phi(x)$, is determined by computing the inverse Mellin transform of m_α . As an example, if the moments m_α are finite for $\alpha < \alpha_c$ and infinite for $\alpha > \alpha_c$, then it is interpreted that $\phi(x)$ asymptotically behaves as the power law, $\phi(x) \propto x^{-1-\alpha_c}$. Although this procedure is not mathematically precise, the correspondence between m_α and $\phi(x)$ is physically plausible, and it is also supported by available exact solutions. This general procedure underlies our ensuing discussion for the asymptotic form of the fragment size distribution.

10.2.4 Scaling solutions to the rate equations

To determine the asymptotic solution to the rate equations, we now substitute the scaling ansatz into eq. (10.9). This separates the mass and time dependence of the rate equation to yield two separate scaling equations for the time dependence of the typical fragment size, and the functional dependence of the fragment size distribution on scaled mass $\xi \equiv x/s$,^[63,64]

$$\omega[2\phi(\xi) + \xi \phi'(\xi)] = -\xi^\lambda \phi(\xi) + \int_\xi^\infty \phi(\eta) \eta^{\lambda-1} b\left(\frac{\xi}{\eta}\right) d\eta, \quad (10.13)$$

$$\dot{s}(t) s(t)^{-(1+\lambda)} = -\omega. \quad (10.14)$$

Here $\omega > 0$ is the separation constant whose value depends on the normalization of m_0 and m_1 , and the overdot denotes the time derivative. From eq. (10.14), the

typical cluster size has the time dependence,

$$s(t) \sim \begin{cases} t^{-1/\lambda}, & \text{for } \lambda > 0 \text{ and } t \rightarrow \infty, \\ e^{-\omega t}, & \text{for } \lambda = 0 \text{ and } t \rightarrow \infty, \\ (t_c - t)^{1/|\lambda|}, & \text{for } \lambda < 0 \text{ and } t < t_c. \end{cases} \quad (10.15)$$

For a smaller value of λ the breakup rate of small clusters plays an increasing role. Therefore the typical size decreases more rapidly for a model with a smaller value of λ . When λ becomes less than zero, the fragmentation process is dominated by the rapid breaking up of the very smallest clusters. This phenomenon is manifested by the fact that the scaling approach predicts that the typical size vanishes in a finite time. This is the signal of the shattering transition, whose existence and properties will be treated in the next subsection. For the present discussion, we consider the case $\lambda > 0$, where scaling can be shown to hold.

To find the asymptotic solution to eq. (10.13), we first convert it to a relation involving the moments of $\phi(\xi)$, by multiplying both sides by ξ^α and integrating over all ξ . After a number of straightforward steps, one obtains a linear recursion relation for the moments of the scaling function,

$$m_{\alpha+\lambda} = \omega \frac{1-\alpha}{L_\alpha-1} m_\alpha, \quad (10.16)$$

where L_α is the α th moment of the reduced breakup kernel

$$L_\alpha = \int_0^1 x^\alpha b(x) dx. \quad (10.17)$$

The explicit dependence on the kernel is *only* contained in L_α . Consequently, the results which follow from this recursion relation will be universal to the extent that they depend only on the homogeneity index that appears in the overall breakup rate and on the limiting behaviour of L_α .

To obtain the time dependence of the moments, we start with the original rate equation and convert it to a moment relation by following exactly the same steps that led to eq. (10.16). This yields the "bare" moment relation,

$$\dot{M}_\alpha = (L_\alpha - 1) M_{\alpha+\lambda}. \quad (10.18)$$

Starting with $M_1(t) = 1$, eq. (10.18) gives $M_{1-\lambda}(t) = (L_{1-\lambda} - 1)t + \text{const.}$, where $L_{1-\lambda} - 1$ is a positive constant. Iterating this process leads to the asymptotic solution,

$$M_{1-k\lambda} \simeq \prod_{j=1}^k (L_{1-j\lambda} - 1) \frac{t^k}{k!} \propto t^k, \quad (10.19)$$

for the discrete set of equidistant index values $1 - k\lambda$. On the other hand, through eq. (10.11) the scaling ansatz gives $m_{1-k\lambda} \propto s^{-k\lambda}$, which is also proportional to t^k . Thus the correct temporal behaviour of the moments is reproduced directly from the scaling ansatz. It is also worth emphasizing that from the theoretical viewpoint, these *negative* moments of the cluster size distribution are the fundamental dynamical quantities that characterize the fragmentation process. They play a role analogous to that of the positive moments of the cluster size distribution in growth phenomena such as aggregation.

From eq. (10.16), we now determine the asymptotic behaviour of m_α and then use the properties of the inverse Mellin transform^[80] to reconstruct the functional form of the scaling function. These details are presented separately for $\alpha \rightarrow \pm\infty$, which from the properties of the Mellin transform, correspond to large and small x , respectively.

10.2.5 Large- x limit

To obtain the moments for large α , choose $\alpha = k\lambda$, with k a positive integer, iterate eq. (10.16), and use $m_0 = 1$. This yields

$$m_{k\lambda} = \omega^{k-1} \prod_{n=1}^{k-1} \frac{n\lambda - 1}{1 - L_{n\lambda}}. \quad (10.20)$$

For large k , the product is dominated by the large- n factors. In this limit, the form of $L_{n\lambda}$ is determined by the behaviour of the breakup kernel $b(x)$ for $x \rightarrow 1$, i.e., in the limit of abrasion. For this limit, consider kernels of the general form $b(x) = b(1) + \mathcal{O}((1-x)^\mu)$, for $x \rightarrow 1$, where $b(1) \geq 0$ and $\mu > 0$ are constants. This means that there is a smoothly varying probability for the production of fragments whose mass is very close to the initial mass, with a limiting probability $b(1)$ corresponding to the case of no breaking. This form for the breaking rate gives

$$L_\alpha = b(1)/\alpha + \mathcal{O}(\alpha^{-(\mu+1)}), \quad (10.21)$$

for large α . Using this result in eq. (10.20), one finds after several straightforward steps,

$$m_\alpha \propto \left(\frac{\omega\alpha}{e}\right)^{\alpha/\lambda} \alpha^{(b(1)-1)/\lambda-1/2}, \quad \text{for } \alpha \rightarrow \infty. \quad (10.22)$$

The non-trivial dependence on the breakup kernel appears only through λ in the controlling factor, $\alpha^{\alpha/\lambda}$, and through $b(1)$ and λ in the correction term, $\alpha^{(b(1)-1)/\lambda}$. This universality of m_α leads, through the inverse Mellin transform, to the following universal expression for the cluster size distribution

$$\phi(x) \sim x^{b(1)-2} \exp(-ax^\lambda) \quad x \rightarrow \infty, \quad (10.23a)$$

where $a = 1/(\lambda\omega)$. By comparing with the scaling ansatz, this gives

$$c(x, t) \propto \exp(-\text{const}.tx^\lambda), \quad (10.23b)$$

which coincides with the asymptotic form of the exact results obtained for binary fragmentation models. The basic conclusion, therefore, is that scaling provides a simple method for obtaining the universal asymptotic form for the cluster size distribution in the large size limit, which agrees with the available exact solutions.

10.2.6 Small- x limit

In the small-mass limit, there is a lesser degree of universality, since the small mass tail is not directly influenced by particles of the typical size. However, there are now just two generic forms for $\phi(x)$, whose applicability depends on whether or not the moments $L_{-\alpha}$ diverge as $\alpha \rightarrow \infty$. Physically, $L_{-\alpha}$ diverging corresponds to the production of a considerable fraction of arbitrarily fine dust in a single breakup event, as might occur in an explosive process.

To obtain $\phi(x)$ in the small- x limit, now requires the behaviour of $m_{-\alpha}$ as $\alpha \rightarrow \infty$. Accordingly, we choose $\alpha = 1 - k\lambda$ in eq. (10.16) and iterate to arrive at the counterpart of eq. (10.20), namely,

$$m_{1-k\lambda} = \omega^{-k} \frac{\prod_{n=1}^k [L_{1-n\lambda} - 1]}{\prod_{n=1}^k n\lambda}. \quad (10.24)$$

In analogy with the case of large positive index values, the k dependence of $m_{1-k\lambda}$ for large k is now determined by the limiting form of $b(x)$ for x near 0, and there appear to be two generic cases for this limiting form. One case is the general situation of kernels which are cut off at small fragment sizes. This corresponds roughly to the case of cleavage, as defined in the previous section. Such a process may be modelled by a kernel with a finite lower limit, x_0 , for the minimum reduction factor in a single breakup event. That is, $b(x) = 0$ for $x < x_0$, with $0 < x_0 < 1$. From eq. (10.17), $L_{1-\alpha}$ has the leading behaviour $x_0^{-\alpha}/\alpha^{1+\mu}$ for large α . Substituting this form into eq. (10.24) yields

$$m_{1-k\lambda} \sim \frac{1}{k!} \frac{1}{(\omega\lambda)^k} x_0^{\lambda+2\lambda+\dots+k\lambda} \propto x_0^{-k^2\lambda/2}. \quad (10.25)$$

Thus the controlling factor of m_α in the large α limit is

$$m_{-\alpha} \sim \exp \left[\frac{-\ln x_0}{2\lambda} \alpha^2 \right] \quad \alpha \rightarrow \infty, \quad (10.26)$$

whose inverse Mellin transform yields the classical log-normal form for the controlling factor of $\phi(x)$,

$$\phi(x) \sim \exp \left[-\frac{\lambda}{2 \ln x_0} (\ln^2 x) \right] \quad (x \rightarrow 0). \quad (10.27)$$

In fact, this expression represents a strict lower bound for the small-mass limit of the cluster-size distribution.

The log-normal form can also be obtained from a simple multiplicative argument which appears to capture some of the essence of repeated fragmentation with a small size cutoff.^[81-85] For such a process, the mass of a given fragment schematically evolves as $x_0 \rightarrow x_1 \rightarrow x_2 \rightarrow \dots \rightarrow x_N$, where the successive reduction factor, $r_k = x_k/x_{k-1}$, is a random variable with a well-behaved distribution. By the central limit theorem, $\log x_N = \sum_{k=0}^N \log r_k$ will be normally distributed, so that x_N will be distributed log-normally. While this naive argument is appealing, it contains the tacit assumption that fragmentation actually proceeds in discrete stages so that each cluster has undergone approximately the same number of breakup events. The essential difference, if any, between the cluster size distribution that arises in this discrete dynamics and from a purely continuous process has not yet been elucidated.

Nevertheless, the log-normal form does appear to describe quite adequately the size distribution in a wide variety of geological situations, such as the distribution of rock sizes in boulder fields and the distribution of particle sizes in soils.^[1-4] Typically, it is found that the log-normal provides a helpful representation of the data whenever the size range of the population varies over several orders of magnitude. For these situations, the log-normal is invoked as a truly fundamental distribution, even though there may be only fair agreement between a given data set and the log-normal form.

A second general class of fragment distributions arises for kernels in which infinitesimal size pieces are produced in a single fragmentation event, as in destructive breaking. Although it is difficult to envision such a process occurring repeatedly, the solution for this class of kernels completes the picture of the possible forms for the small mass tail of the fragment size distribution. The absence of a cutoff in the breakup kernel is typified by the power-law form $b(x) \sim x^\nu$ for small x . Thus from eq. (10.16), m_α diverges whenever L_α diverges. This divergence 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. (10.16) to give

$$m_\alpha \simeq L_\alpha \frac{m_{\alpha_c+\lambda}}{\omega(1-\alpha_c)} \propto L_\alpha. \quad (10.28)$$

Since m_α is proportional to L_α , it follows that $\phi(x)$ coincides with $b(x)$. That is $\phi(x)$ has the power-law form

$$\phi(x) \sim x^\nu, \quad \text{as } x \rightarrow 0. \quad (10.29)$$

Such a power-law behaviour is seen in a variety of fragmentation experiments.^[52,55,62]

Thus for a kernel with no small size cutoff, the limiting form of $\phi(x)$ now decays as $\exp(-\nu \ln x)$, which should be compared to the more rapidly varying log-normal bound, $\exp[-c(\ln x)^2]$ for a kernel with a small size cutoff. Eqs. (10.23), (10.27) and (10.29) provide the asymptotic behaviours of $\phi(x)$ for an encompassingly wide class of breakup kernels. These differing forms for the fragment size

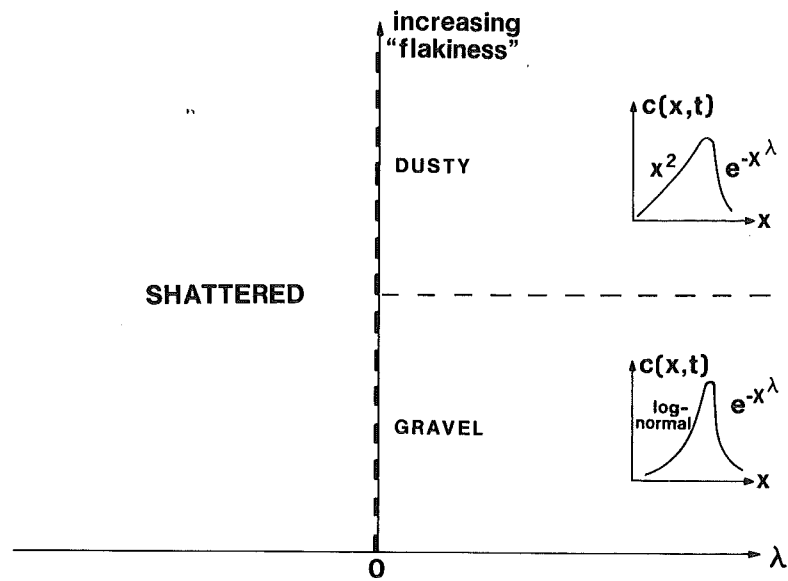


Figure 10.4 Phase diagram for linear fragmentation in the plane defined by the homogeneity index λ and a loosely-defined parameter, the "flakiness" of the relative breakup rate. Small flakiness corresponds to a vanishingly small probability of small flakes being produced in a single breakup event, i.e., a small-size cutoff in $b(x)$, while large flakiness corresponds to the opposite limit of a power-law tail in $b(x)$. In the phase plane there is a "shattered" phase for $\lambda < 0$, while for $\lambda \geq 0$ there is a "gravel" phase for small flakiness and a "dusty" phase for large flakiness. The fragment size distributions corresponding to these latter two phases are sketched (from ref. [64]).

distribution may be considered as corresponding to different “phases” of fragment products, as illustrated by the “phase diagram” of fig. 10.4.

10.2.7 Breakdown of scaling and shattering transition for $\lambda < 0$

For $\lambda < 0$, conventional scaling solutions to the rate equations do not exist.^[51,60,64] Accompanying this anomaly is the intriguing phenomenon of “shattering”, in which mass is “lost” to a phase of zero mass particles as the fragmentation progresses. Thus $\lambda = 0$ is the dividing point which separates models for which conventional scaling exists from models which undergo shattering. Although shattering may be a mathematical pathology, since small fragments are generally more difficult to break than large clusters at fixed energy density, there are analogies with the inverse and physical process of gelation.^[68–72] Furthermore, the shattering phenomenon reveals important and intriguing features about the structure of the solutions to the rate equations, which can be usefully studied by exact solutions and by scaling approaches.

The physical manifestation of shattering is mass loss in the system, $\dot{M}_1 < 0$ (more specifically, mass loss from the population of finite size clusters). As first shown generally by Filippov,^[51] and by McGrady and Ziff^[60] for specific fragmentation models, this occurs when $\lambda < 0$. We now give a simple argument to show that $\lambda < 0$ is the necessary and sufficient condition for shattering in a homogeneous fragmenting system.^[64] To locate the shattering transition, we write eq. (10.18) for $\alpha = 1 + \epsilon$,

$$\dot{M}_1 = (L_{1+\epsilon} - 1) M_{\lambda+1+\epsilon}. \quad (10.30)$$

As $\epsilon \rightarrow 0^+$, L_α approaches unity from below (mass conservation), so that \dot{M}_1 can be nonzero only if $M_{\lambda+1+\epsilon}$ diverges as $\epsilon \rightarrow 0$. Without loss of generality, suppose that the largest initial cluster mass is unity, so that there will be no cluster with a mass larger than unity for $t > 0$. Then M_α is non-decreasing as α decreases, at any 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 $\lambda < 0$.

To show that $\lambda < 0$ is also a sufficient condition for shattering, let us assume the converse and derive a contradiction. For $\lambda < 0$, eq. (10.18) gives

$$\dot{M}_{1+|\lambda|} = (L_{1+|\lambda|} - 1) M_1. \quad (10.31)$$

Under the assumption of no shattering, M_1 is fixed, so that the right-hand side is a negative constant. This implies that $M_{1+|\lambda|}$, and consequently $c(x, t)$ would vanish at a finite time. On the other hand, we know directly from the rate equations that $c(x, t)$ must decay exponentially, or slower, in time for any x . This contradiction implies that a necessary and sufficient condition for shattering to occur is $\lambda < 0$.

Some of the peculiar features of the cluster size distribution can already be seen from the exact solution of the rate equations for binary breakup in which the homogeneity exponent equals zero. This corresponds to a system on the borderline

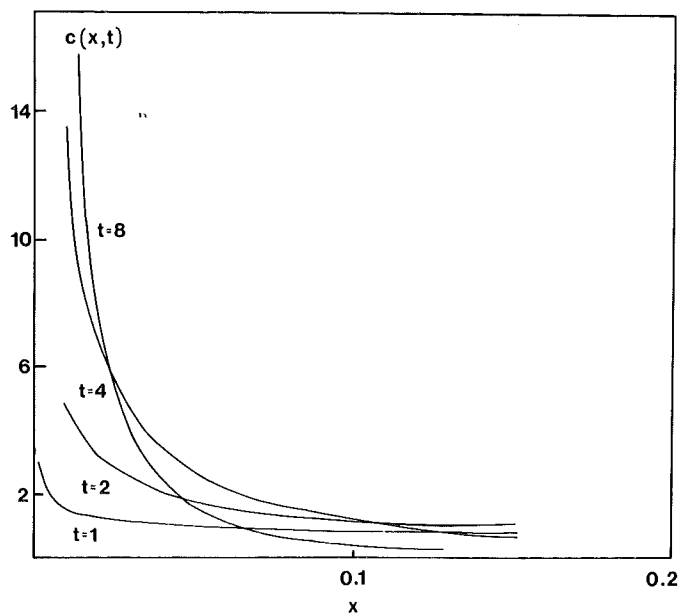


Figure 10.5 Sketch of the size dependence of the exact fragment size distribution at fixed times of binary breakup when $F(x, y) \sim (x + y)^{-1}$ (eq. 10.32). The singular behaviour in the small-size limit should be compared with the regular small-size behaviour for fragmenting systems with positive homogeneity exponent (fig. 10.3b).

between scaling and shattering. For $F(x, y) \propto (x + y)^{-1}$, the exact solution for the cluster size distribution in the limit $\lambda \rightarrow 0$ is,^[58]

$$c(x, t) = e^{-t} \delta(x - l) + \frac{2t e^{-t}}{l} \sum_{n=0}^{\infty} \frac{[2t \ln(l/x)]^n}{n!(n+1)!}, \quad (10.32)$$

and the corresponding moments of this cluster size distribution are,

$$M_n(t) = l^n \exp[(1 - n)t/(1 + n)]. \quad (10.33)$$

Thus the total number of fragments, $M_0(t)$, grows exponentially in time, in contrast to the power-law growth for $M_0(t)$ in the non-shattering regime. A sketch of $c(x, t)$ versus x for several values of t is shown in fig. 10.5. In the small mass limit, a singularity in $c(x, t)$ develops which signals the explosive growth of very small clusters. This singularity nevertheless makes a finite contribution to the total number of clusters.

It is also possible to develop an alternative scaling formulation for $c(x, t)$ which accounts for basic features of the fragment size distribution in the shattering

regime. To justify this new scaling ansatz, recall that the conventional scaling solution for $\phi(x)$ (eq. (10.23)) yields $c(x, t) \sim e^{-x^\lambda t}$. The coefficient of t in the exponential is a decreasing function of x for $\lambda > 0$, so that smaller clusters decay more slowly. On the other hand, for $\lambda < 0$, one can show directly from the rate equations that $c(x, t) \sim e^{-t^{-|\lambda|} t}$ for the monodisperse initial condition $c(x, t = 0) = \delta(x - l)$, where l is the initial mass of the fragments. That is, the coefficient of t in the exponential is *fixed*, or equivalently the typical mass is *time-independent*. This anomaly reflects the fact that once the first few small clusters are produced, they react so quickly that the initial mass remains behind as a virtually non-reactive residue, and this residue determines the typical size. This is similar to the behaviour in the inverse process of gelation, where small clusters remain behind as the non-reactive sol phase in the presence of a very few, highly-reactive, large macromolecules.

These arguments, together with an examination of the conventional scaling ansatz and exact solutions in the shattering regime,^[64] lead to the following form for the long-time behaviour of $c(x, t)$ when $\lambda < 0$,

$$c(x, t) = T(tl^{-|\lambda|})\phi(x/l), \quad (10.34)$$

where $T(u)$ is a scaling function. This resembles the conventional scaling ansatz, with $s(t)$ being replaced by l in the scaling function ϕ . Since we have shown that $c(x, t)$ has the time dependence $e^{-t^{-|\lambda|} t}$, it now remains to determine the function $\phi(x)$ to complete the description of the fragment size distribution. By a careful examination of the moment equation for $\lambda < 0$, it is possible to show that $\phi(x)$ has the universal form $x^{|\lambda|-2}$. Thus we conclude that in the shattering regime, $c(x, t)$ has the universal behaviour

$$c(x, t) \sim e^{-t^{-|\lambda|} t} x^{|\lambda|-2}. \quad (10.35)$$

This functional form can also be justified by a very simple argument which is based on the physical picture that most of the mass in the system remains with the nearly-constant initial mass residue. If the concentration of this residue was strictly constant, then the dynamics of the system could be described by the rate equation (10.3), augmented by the presence of a constant *source* of clusters of mass unity. For this modified rate equation, the moment relation becomes^[64]

$$m_{\alpha-|\lambda|} \simeq \frac{1}{1 - L_\alpha} \quad (\alpha \gtrsim 1 - |\lambda|). \quad (10.36)$$

This turns out to coincide with a detailed calculation for moments of the cluster size distribution for irreversible fragmentation in the $\lambda < 0$ regime. Thus we conclude that for $\lambda > 0$, clusters of all sizes participate substantially in fragmentation, leading naturally to scaling behaviour. However for $\lambda < 0$, only the very smallest clusters fragment at any appreciable rate. The singular aspects of the fragment size

distribution are influenced by this population of vanishingly small clusters, while the scaling portion of the distribution is determined by the nearly non-reactive residue of initial-mass clusters. The features of the cluster size distribution also occur in the inverse process of gelation.^[68-72]

10.2.8 Comparison with comminution experiments

As mentioned at the outset of this chapter, there are many examples of continuous fragmentation. Indeed, the preparation of a dispersed product by the grinding of a coarse feed is an underlying theme in most mining engineering and powder technology applications.^[11-15] The grinding or comminution of minerals by milling machinery, in particular, provides an excellent laboratory for the comparison of theory and experiments. Experimental conditions can be easily controlled, and one has a reasonable physical picture of the microscopic breakup processes that are involved in grinding. The detailed comparison between theory and experiments therefore seems to be best developed in the literature on comminution. In the comminution of ores, the important practical question to address is the relation between the energy input to the system and the degree of size reduction of the ore. More precisely, for a given distribution of feed and for a fixed grinding time, the issue of practical relevance is the size distribution of the ground material.

In comminution by ball or rod milling, for example, the initial feed is placed in a cylindrical container, which is typically of the order 10^2 cubic meters in volume, and spins at a rate that is typically of the order of 10^2 revolutions per minute.^[11] The grinding action is accomplished by the presence of a very hard milling medium, such as steel balls or steel rods. As the container spins, the collision of the ground material with the steel is the primary mechanism for size reduction. The grinding time is typically of the order of minutes to many hours, depending on the degree of size reduction needed. A wide variety of geometries and milling processes have been devised, depending on the specific application that is required. The large degree of mixing inherent in a tumbling grinding mill suggests that material is homogeneously distributed in the mill. (However, for steady-state operation, mills have been designed to produce size segregation, with smaller fragments close to the output end.^[11]) If spatial homogeneity does apply, then one of the basic assumptions of the rate equations is satisfied. Thus for milling, at least, the mean-field rate equations do provide an appropriate starting point.

On the basis of these types of comminution experiments, a number of basic conclusions have been drawn. First, for processes such as ball or rod milling, the fragmentation process is linear to a high degree of accuracy.^[12,55] This linearity has been tested by verifying that the concentration of material within a given size range is depleted at an exponential rate. This can be accomplished by studying the subpopulation of the largest initial clusters, or alternatively, following the evolution of a radioactively marked subpopulation of smaller clusters in the initial feed. Experiments on many types of ground media also indicate that the overall breakup rate of a cluster of size x , $a(x)$, is a decreasing function of x , whose precise form

depends on the material being ground and on the milling conditions. For one class of examples where careful measurements have been performed, $a(x)$ is found to vary as x^λ , with $\lambda \simeq 0.3 - 0.4$ for materials such as coal, limestone, and cement clinker in ball milling conditions.^[52,55,61] In addition, these experiments also suggest that the relative breakup rate, $f(x|y)$, has a power-law behaviour, $f(x|y) \sim (x/y)^\beta$ for a wide range of conditions. As shown in section 10.3, the corresponding fragment size distribution should therefore have power-law tail at the small-size limit. Experimentally there does appear to be a relatively universal behaviour, with the cumulative mass distribution of mass less than x varying as x^γ , with γ in the range 0.6 to 0.9.

Thus in strongly mixed milling media, the connection between the theoretical treatment of the rate equations and measurements is quite reasonable. For a wide class of systems, it is even possible to extract a value for the homogeneity exponent of the overall breakup rate, and also the nature of the relative breakup rate from the apparent power-law small size tail of fragment sizes.

10.3 Discussion and outlook

Fracture and fragmentation underlie a wide variety of dynamical processes. In this chapter, I have attempted to outline the basic features of continuous fragmentation and the underlying, relevant aspects of instantaneous fracture. It has been found that over a wide range of energy input to the system, it is generally harder to break up small clusters than large clusters. This corresponds to an overall breakup rate which is a decreasing function of the mass, in the rate equations for continuous fragmentation. For the *distribution* of fragment sizes that are produced in a single instantaneous fragmentation event, there is considerable data which indicates that for many materials and experimental situations, the cumulative mass fraction which is smaller than mass x varies as x^α , with α generally close to unity. This corresponds to the relative abundance of fragments of size x varying as $x^{2-\alpha/3}$ (eq. (10.1)). This power law form holds over several decades in the best experimental situations, but clearly, there must be a cutoff in which a distribution at the very smallest scale. Such a power-law form translates to a relative breakup rate that does not have a small-size cutoff in the rate equations, a situation in which a small-size cutoff must eventually play a prominent role.

A general treatment of continuous fragmentation has been given within the framework of the rate equations. This is an approach of a mean-field character, as fluctuations in the spatial positions of the clusters and in cluster shape are ignored. Both of these approximations are quite drastic, and very little is known about how to account for these fluctuations in a realistic and tractable manner. One does expect that the rate equations will provide the correct asymptotic behaviour for systems of sufficiently high spatial dimensionality, and that the mean-field description will break down below a critical dimensionality d_c . Presently, there is no understanding of how to determine this upper critical dimension. In contrast, for

the inverse process of irreversible aggregation, there are well-established methods for determining d_c , and it is known that $d_c = 2$ for systems where the cluster reactivity is mass independent,^[86,87] and that d_c may be infinite for models in which the reactivity is a strongly increasing function of mass.^[88] In fragmentation, however, there is no obvious universal transport or mixing mechanism, akin to Brownian motion in aggregating systems, with which one can begin to estimate the role of fluctuations within a typical correlation volume. Thus it is an open question of how to assess the degree of validity of the rate equations for describing the kinetics of continuously fragmenting systems.

Nevertheless, the rate equations do provide a simple and comprehensive treatment for the kinetics of continuous fragmentation. Within this approach, the microscopic details of breakup events are accounted for by the overall breakup rate, which specifies the likelihood for a fragment of a given mass to break (either by an external agent or by collisions), and the relative breakup rate, which gives the size distribution of products from a single breakup event. These fragmentation kernels can be determined empirically for a wide range of systems. For many functional forms for the breakup rates, it is possible to solve the rate equations in closed form. A more general, yet very simple approach is based on scaling, in which it is assumed that the fragment size distribution depends on mass and time only through the scaling combination of the ratio of the mass to the typical mass. For a homogeneous fragmenting system, scaling solutions exist when the homogeneity index, λ , of the overall breakup rate is positive. Within the scaling regime, the value of λ is the crucial parameter which characterizes the large-mass behaviour of the cluster size distribution. Under very general conditions, the large mass tail of the distribution is found to decay as $e^{-t\omega^\lambda}$. In the small-mass limit, the cluster size distribution is determined by whether or not the relative breakup rate is cut off in the small-size limit. With a cutoff, the cluster size distribution has a log-normal tail, while in the opposite case, there is a power-law tail of very small dust-like particles.

When $\lambda < 0$, a shattering transition occurs in which mass is lost to a dust phase consisting of an infinite number of zero mass particles. It is possible to obtain asymptotic dynamical behaviour through a scaling ansatz if one takes the initial mass as the characteristic mass scale of the system. Owing to the rapid breakup of very small clusters, the initial mass decays away extremely slowly, so that this component of the distribution acts as a steady source for the remainder of the fragment size distribution. This intuitive picture accords with exact solutions of fragmenting systems with $\lambda < 0$.

While the rate equations are useful for treating a wide range of continuous fragmentation phenomena, there are interesting questions of potential experimental relevance that should be addressed. At a simple-minded level, the validity of a purely linear theory can be questioned in situations as violent as rock crushing and in many other fragmentation phenomena. In rock grinding experiments, deviations from the predictions of linear rate equations are found to occur in various

cases.^[55] Both non-linear and non-local effects can be expected to play some role in high energy fragmentation processes. When breakup is driven by high pressures, there can be a transfer of stress across many fragments. When fragments possess considerable kinetic energy, non-linear collisional effects may play a substantial role.^[38] Some of these features are embodied in *autogeneous* milling,^[11] where the breakup of fragments is caused *only* by the material undergoing comminution. A satisfactory theoretical description has not yet been developed for this intriguing non-linear process. A natural starting point worth developing is to consider the rate equations with either time-dependent or concentration-dependent breakup rates. Autogeneous milling also shares some features with an idealized collision-induced fragmentation model, in which fragmentation is driven only by binary collisions between fragments.^[64,89] In the rate equations for this process, fragmentation events are driven both by an intrinsic breakup rate and by the rate at which two fragments meet. The latter is described by the time rate of change of the concentration $\dot{c}(x, t)$ being proportional to the square of the overall density.^[64] This collision-induced model exhibits a rich phenomenology, whose solution may provide a starting point for treating autogeneous fragmentation.

Another potential example of a system undergoing non-linear fragmentation is the asteroid belt, if collisions between asteroids are the primary mechanism that lead to continued breakup events. In addition to the apparent non-linearity, there is inhomogeneity and possible self-organization in the spatial and velocity distributions of the asteroids. In view of these facts, one can reasonably doubt the applicability of a rate equation approach to discuss the size distribution of asteroids. Nevertheless, an early rate equation approach^[8] predicted that the density of fragments of mass x varies approximately as $x^{-1.8}$. This was in good agreement with the observational data of that time, but current data reveals that it is inadequate to view asteroids as a single homogeneous population.^[10] It remains to be established whether a rate equation approach can capture the essential features of asteroid dynamics and fragmentation.

In a more practical vein, it is important to develop approximations which can take inhomogeneities of various types into account. While the assumption of perfect mixing is most likely appropriate for ball milling comminution, perfect mixing is clearly inadequate in many geophysical situations, where fragments may remain in fixed spatial positions throughout the breakup process. Attempts to account for this type of spatial inhomogeneity have been attempted, but primarily at a qualitative level. For the phenomenon of the formation of fragments (gouge) in an earthquake fault, a "constrained comminution" model has been introduced,^[90] in which it is postulated that in a system of fragments with fixed relative positions, fragmentation is most likely to occur between neighbouring fragments of the same size. This model is based on the intuition that neighbouring fragments of dissimilar sizes will tend to "nest", leading to a screening of tensile forces. This model leads also to the question of how the rate of fragmentation depends on cluster shape.

Whether this shape dependence can be accounted for by averaging over fragment shapes has not been seriously considered. This potential shape dependence also appears to be ignored in the available experimental fragmentation data. Also noteworthy is a related constrained fragmentation model of Derrida and Flyvbjerg,^[91] which has a very different physical application in mind. In their model, there is binary fragmentation of the unit interval, in which only one of the two product fragments is available for continued breakup. This model was introduced to describe the evolution of the phase space of a disordered system, such as a spin glass, as the temperature is lowered. This model is particularly intriguing for the rich singularity structure exhibited by the fragment size distribution.

In conclusion, fracture and fragmentation encompasses a wide range of dynamic phenomena. The theoretical treatment of linear fragmentation processes is fairly well-developed. However, there are many situations where non-linearity, and inhomogeneities of various types will play an important role in the breakup process. The understanding of the dynamics for these types of systems should pose rich areas for new research.

I wish to thank Zheming Cheng for a most enjoyable collaboration on some of the work reported here. This work has been supported in part by grant #DAAL03-89-K-0025 from the Army Research Office. This financial support is gratefully acknowledged.

References

1. E.C. Dapples, *Basic Geology for Science and Engineering* (Wiley, New York, 1959)
2. *Models of Geologic Processes; An Introduction to Mathematical Geology*, Ed. P. Fenner (American Geological Institute, Washington, 1969)
3. P. Habib, *Soil and Rock Mechanics* (Cambridge University Press, Cambridge, 1983)
4. T. H. Wu, *Soil Mechanics* (Allyn and Bacon, Boston, 1966)
5. See, e.g., *Geology and Geophysics of the Moon*, Ed. G. Fielder (Elsevier, Amsterdam, 1972)
6. J. Guest, *Geology of the Moon* (Wykeham Publications, London, 1977)
7. W. K. Hartmann, *Icarus* **10**, 210 (1969)
8. J. S. Dohnanyi, *J. Geophys. Res.* **74**, 2531 (1969)
9. C. J. Cunningham, *Introduction to Asteroids* (Willmann-Bell, Inc., Richmond, VA, 1988)
10. See, e.g., the collection of articles in *Asteroids* Ed. T. Gehrels (University of Arizona Press, Tucson, 1979)
11. E. G. Kelley and D. J. Spottiswood, *Introduction to Mineral Processing* (Wiley, New York, 1982)
12. L. G. Austin, R. R. Klimpel, and P. T. Luckie, *Process Engineering of Size Reduction: Ball Milling* (Society of Mining Engineers, New York, 1984)

13. L. G. Austin and R. R. Klimpel, *Ind. Eng. Chem.* **56**, 18 (1964)
14. E. J. Pryor, *Mineral Processing* (Elsevier, Amsterdam, 1965)
15. A. J. Lynch, *Mineral Crushing and Grinding Circuits* (Elsevier, Amsterdam, 1977)
16. H. Mark and R. Simha, *Trans. Faraday Soc.* **35**, 611 (1940)
17. E. Montroll and R. Simha, *J. Chem Phys.* **8**, 721 (1940)
18. P. J. Blatz and A. V. Tobolsky, *J. Phys. C* **49**, 77 (1945)
19. R. Simha, L. A. Wall, and P. J. Blatz, *J. Polymer. Sci.* **5**, 615 (1950)
20. H. H. Jellinek and G. White, *J. Polymer. Sci.* **6**, 745 (1951)
21. A. M. Basedow, K. H. Ebert, and H. J. Ederer, *Macromolecules* **11**, 774 (1978)
22. R. M. Ziff and E. D. McGrady, *Macromolecules* **19**, 2513 (1986)
23. C. A. Sundback, J. M. Beér, and A. F. Sarofim, *Proc. 20th Int. Symp. on Combustion* (The Combustion Institute, Pittsburgh, 1985)
24. G. Daccord and R. Lenormand, *Nature* **325**, 41 (1986)
25. D. Dunn-Rankin and A. R. Kerstein, *Combust. and Flame* **69**, 193 (1987)
26. A. R. Kerstein and B. F. Edwards, *Chem. Engr. Sci.* **42**, 1629 (1987)
27. M. Sahimi and T. T. Tsotsis, *Phys. Rev. Lett.* **59**, 888 (1987)
28. R. Shinnar, *J. Fluid Mech.* **10**, 259 (1961)
29. D. Ramkrishna, *Chem. Engr. Sci.* **29**, 987 (1974)
30. L. A. Tavlarides and M. Stamatoudis, *Adv. Chem. Eng. Sci.* **11**, 199 (1981)
31. P. Becher (ed.), *Encyclopedia of Emulsion Technology, vol. 1* (Marcel Dekker, New York, 1983)
32. A. P. Siebesma, R. R. Tremblay, A. Erzan, and L. Pietronero, *Physica A* **156**, 613 (1989)
33. See, e.g., D. L. Turcotte, *J. Geophys. Res.* **91**, 1921 (1986), for a recent review
34. B. R. Lawn and T. R. Wilshaw, *Fracture of Brittle Solids* (Cambridge University Press, Cambridge, 1975)
35. S. M. Weiderhorn, *Ann. Rev. Mater. Sci.* **14**, 373 (1984)
36. R. W. Davidge *Mechanical Behavior of Ceramics, Cambridge Solid State Sciences Series* (Cambridge University Press, Cambridge, 1979)
37. See, e.g. the collection of articles in *Fracture Vol. I - VII*, H. Liebowitz ed., (Academic Press, New York, 1984)
38. See e.g., *Fragmentation, Form and Flow in Fracture Media*, R. Engelman and Z. Jaeger eds., *Annals of the Israel Physical Society*, vol. 8 (Adam Hilger, Bristol, England, 1986)
39. P. Rosin and E. Rammner, *J. Inst. Fuel* **7**, 29 (1933)
40. R. Schuhmann, Jr., *Trans. AIME/SME* **217**, 22 (1960)
41. J. J. Gilvarry, *J. Appl. Phys.* **32**, 391 (1961)
42. J. J. Gilvarry and B. H. Bergstrom, *J. Appl. Phys.* **32**, 400 (1961)
43. J. J. Gilvarry and B. H. Bergstrom, *Trans. AIME/SME* **193**, 484 (1961)
44. A. M. Gaudin and T. P. Meloy *Trans. AIME/SME* **223**, 40 (1962); **223**, 43 (1962)
45. R. R. Klimpel and L. G. Austin *Trans. AIME/SME* **232**, 88 (1965)

46. A. Fujiwara, G. Kamimoto, and A. Tsukamoto, *Icarus* **31**, 277 (1977)
47. M. A. Lange, T. J. Ahrens, and M. B. Boslough, *Icarus* **58**, 383 (1984)
48. C. J. Allègre, J. L. Le Mouel, and A. Provost, *Nature* **297**, 47 (1982)
49. R. F. Smalley, Jr., D. L. Turcotte, and S. A. Solla, *J. Geophys. Res.* **90**, 1894 (1985)
50. D. L. Turcotte, R. F. Smalley, Jr., and S. A. Solla, *Nature* **90**, 1894 (1985)
51. A. F. Filippov, *Theory Probab. and its Appl.* (Engl. Transl.) **4**, 275 (1961)
52. D. D. Crabtree, R. S. Kinasevich, A. L. Mular, T. P. Meloy, and D. W. Fuerstenau, *Trans. AIME/SME* **229**, 201 (1964)
53. L. G. Austin, *Powder Tech.* **5**, 1 (1971)
54. V. K. Gupta and P. C. Kapur, *Powder Tech.* **12**, 175 (1975)
55. L. Austin, K. Shoji, V. Bhatia, K. Savage, and R. Klimpel, *Ind. Eng. Chem. Process Des. Dev.* **15**, 187 (1976)
56. L. G. Austin and P. Bagga, *Powder Tech.* **28**, 83 (1981)
57. L. G. Austin, P. Bagga and M. Celik, *Powder Tech.* **28**, 235 (1981)
58. R. M. Ziff and E. D. McGrady, *J. Phys. A* **18**, 3027 (1985)
59. R. M. Ziff and E. D. McGrady, *Macromolecules* **19**, 2513 (1986)
60. E. D. McGrady and R. M. Ziff, *Phys. Rev. Lett.* **58**, 892 (1987)
61. T. W. Peterson, M. V. Scotto, and A. F. Sarofim, *Powder Tech.* **45**, 87 (1985)
62. T. W. Peterson, *Aerosol Sci. Tech.* **5**, 93 (1986)
63. Z. Cheng and S. Redner, *Phys. Rev. Lett.* **60**, 2450 (1988)
64. Z. Cheng and S. Redner, *J. Phys. A* (1990) to appear
65. B. F. Edwards, M. Cai, and H. Han, preprint
66. M. Cai, B. F. Edwards, and H. Han, preprint
67. S. K. Friedlander and C. S. Wang, *J. Colloid Interface Sci.* **22**, 126 (1966)
68. R. M. Ziff, *J. Stat. Phys.* **23**, 241 (1980)
69. M. H. Ernst, E. M. Hendriks, and R. M. Ziff, *J. Phys. A* **15**, L743 (1982)
70. F. Leyvraz and H. R. Tschudi, *J. Phys. A* **14**, 3389 (1981); *J. Phys. A* **15**, 1951 (1982)
71. E. M. Hendriks, M. H. Ernst, and R. M. Ziff, *J. Stat. Phys.* **31**, 519 (1983)
72. M. H. Ernst, R. M. Ziff, and E. M. Hendriks, *J. Colloid Interface Sci.* **97**, 266 (1984)
73. T. Vicsek and F. Family, *Phys. Rev. Lett.* **52**, 1669 (1984)
74. M. Kolb, *Phys. Rev. Lett.* **53**, 1653 (1984)
75. P. G. D. van Dongen and M. H. Ernst, *Phys. Rev. Lett.* **54**, 1396 (1985)
76. K. Kang, S. Redner, P. Meakin, and F. Leyvraz, *Phys. Rev. A* **33**, 1171 (1986)
77. A. A. Griffith *Phil. Trans. Roy. Soc., Ser. A* **221**, 163 (1920-1921)
78. H. Rumpf, *Powder Tech.* **7**, 145 (1973)
79. Y. Oka and W. Majima, *Can. Met. Q.*, **9**, 429 (1970)
80. See, e.g., J. Mathews and R. L. Walker, *Mathematical Methods of Physics* (Addison-Wesley, Reading, Massachusetts, 1970)
81. A. N. Kolmogorov, *Doklady Akad. Nauk. SSSR* **31**, 99 (1941)
82. P. R. Halmos, *Ann. Math. Stat.* **15**, 182 (1944)

83. B. Epstein, *J. Franklin Inst.* **244**, 471 (1947)
84. See e.g., J. Aitchison, *The lognormal distribution* (Cambridge University Press, London, England, 1957), and references therein
85. See e.g., E. W. Montroll and M. F. Shlesinger, in *Nonequilibrium Phenomena II: From Stochastics to Hydrodynamics*, eds. J. L. Lebowitz and E. W. Montroll (North-Holland, Amsterdam, 1984)
86. S. Redner and K. Kang, *Phys. Rev. A* **30**, 2833 (1984)
87. L. Peliti, *J. Phys. A* **19**, L365 (1986)
88. P. G. D. van Dongen, *Phys. Rev. Lett.* **63**, 1281 (1989)
89. See also R. C. Srivastava, *J. Atmos. Sci.* **39**, 1317 (1982), for a particular limit of nonlinear fragmentation
90. C. Sammis, G. King, and R. Biegel, *PAGEOPH* **125**, 777 (1987)
91. B. Derrida and H. Flyvbjerg, *J. Phys. A* **20**, 5273 (1987)