

Fragmentation-inactivation models with mass loss

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We consider one-dimensional discrete models for dynamic fragmentation with mass consumption. The fragmentation cascade is randomly interrupted by inactivation of fragments. Exact solutions are obtained. The scaling regimes for the average number of fragments $n(s, t)$ with mass s at time t are investigated for short and long times. We also study numerically the fluctuations in the size distribution of inactivated sites (intermittency analysis).

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I. INTRODUCTION

Fragmentation processes appear in several areas of physics. They can be essentially classified as (i) *processes with mass conservation* such as grinding and crushing a mineral [1,2], polymer degradation (mechanical, thermal, and radiation induced) [3–6], and droplet breakup [7–9]; (ii) *processes with mass loss* such as oxidation, melting, and sublimation or dissolution of the exposed surface of a porous solid [10]. The physics of the processes with constant mass has been considered in the framework of linear rate equations for continuous systems [11]. In particular, an unexpected loss of mass to zero-mass particles (*shattering transition*) has been found [12] and a scaling theory has been developed [13]. On the other hand, processes with mass loss have been considered in continuous and discrete systems. A scaling theory [14] has been extended for linear continuous fragmentation processes. Discrete models have been studied by means of computational simulations [15,16] and by analytical approaches to rate equations [17].

Recently, intermittency has been found in particle and nuclear multifragmentation [18]. In order to explain the large nonstatistical fluctuations of the fragment-size distribution of the breakup of high-energy nuclei in nuclear emulsion, a model of binary fragmentation has been proposed [19]. In addition to the usual gain (breakup) term, a loss term (randomly inactivating the fragmentation cascade) has been introduced. An intermittent behavior of the fluctuations in the cluster-size distribution has been found for asymptotic times.

In this work we consider some one-dimensional discrete models for fragmentation with mass consumption and inactivation. The system to be consumed is modeled by clusters of occupied sites in a lattice. The attack of the external fields (chemical or physical) is described by site consumption rates. We assume that each occupied site of the lattice has the same probability of being consumed. During the process of fragmentation a cluster of s sites is inactivated randomly with a given rate. This means that this cluster is frozen and its sites cannot be consumed anymore. It represents, in some sense, a non-

reactive portion of the system. For long times, only inactivated clusters can be present. The dynamical process of inactivation can simulate in an easy way a heterogeneous system, in which the several portions have different reactivity. Then each portion can be consumed in different times. For a given time scale, some clusters are consumed and others not. The inactivated clusters represent the latter. These models can be applied to study the distribution of ash particles sizes resulting from the combustion of porous media. This problem is very important for ambiental pollution. Now, an inactivated cluster represents the ash produced in the consumption of these sites. After the total consumption of the porous media, the ash particle size distribution can be evaluated. We are interested in exact results of the rate equations for the time-dependent size distribution of fragments and its fluctuations. We found no intermittent behavior for long times.

This paper is organized as follows. In the following section we describe the models with consumption and inactivation and we present the interesting macroscopic quantities and the rate equations for the average number of fragments with s sites at time t . In Sec. III we present the analytical results for the model with inactivation independent of the size of the cluster. The model with inactivation directly proportional to the size of the cluster is presented in Sec. IV. We discuss the characterization of intermittency and present our numerical simulations in Sec. V. Finally, in Sec. VI we present some concluding remarks.

II. MODELS AND RATE EQUATIONS

Consider an open one-dimensional lattice with a fraction of its sites occupied by unit masses. Through attack of the external field, an occupied site can lose its mass and become empty. This happens at the rate a independent of the position of the site. A fragment is a cluster of occupied sites of the lattice. It is characterized only by its size (or mass), which is defined by the total number of sites belonging to the cluster. At time $t = 0$ there is

only a cluster of s_0 occupied sites. The initial fragment can be divided in two new fragments when a site is consumed. These two fragments can generate four new ones and as long as this process evolves, a fragmentation cascade is established. It can be interrupted by a randomly inactivation of clusters, since the sites of an inactivated fragment cannot be consumed. We consider that each cluster of size s is inactivated with a rate b_s . Now we have two kind of occupied sites: those that can be consumed (active sites) and those that are frozen (inactive sites). The fragmentation process terminates when all sites are inactive or consumed.

All properties are derived from the microscopic quantities $n(s, t)$ and $m(s, t)$, the average number at time t of clusters with s active and inactive sites, respectively. Note that $n(s, t)$ [or $m(s, t)$] is proportional to the probability that a site of the lattice belongs to an active (or inactive) fragment of size s at time t . It is easy to write the coupled linear equations governing the dynamics of $n(s, t)$ and $m(s, t)$

$$\frac{d}{dt} n(s_0, t) = -[s_0 a + b_{s_0}] n(s_0, t) , \quad (1)$$

$$\frac{d}{dt} n(s, t) = -[s a + b_s] n(s, t) + \sum_{k=s+1}^{s_0} 2 a n(k, t) \quad \text{for } s < s_0 , \quad (2)$$

$$\frac{d}{dt} m(s, t) = b_s n(s, t) \quad \text{for all } s . \quad (3)$$

Let us discuss the definition of some macroscopic quantities. The number of active and inactive fragments are defined by $N_n(t) = \sum_{s=1}^{s_0} n(s, t)$ and by $N_m(t) = \sum_{s=1}^{s_0} m(s, t)$. Then, the total number of fragments is $N(t) = N_n(t) + N_m(t)$. The total mass $M(t)$ existing at time t is equal to the number of occupied sites at that time. So we have that $M(t) = \sum_{s=1}^{s_0} s [n(s, t) + m(s, t)]$. Note that $M(t) = M_n(t) + M_m(t)$, where $M_n(t)$ is the active mass and $M_m(t)$ is the inactive mass. Another interesting macroscopic quantity is the average fragment size, which can be defined as $\langle s \rangle = M(t)/N(t)$. A similar definition is used for the average size of the active (inactive) fragments. The dynamic equations for all these macroscopic quantities are readily obtained by summing Eqs. (1)–(3) for all s .

Equations (1) and (2) for $n(s, t)$ can be viewed as a set of s_0 first-order linear differential equations with constant coefficients and can be rewritten as

$$\frac{d}{dt} \vec{n}(t) = X \vec{n}(t) , \quad (4)$$

where \vec{n} is the s_0 -dimensional vector $(n(1, t), n(2, t), \dots, n(s_0, t))$ and X is an $s_0 \times s_0$ matrix [20]. Since the matrix X is triangular, its eigenvalues are the diagonal elements $\lambda_i = -i[a + b_i]$. The corresponding eigenvectors $v^i = (x_1^i, x_2^i, \dots, x_i^i, 0, \dots, 0)$ can be recursively determined through

$$x_j^i = \frac{1}{\lambda_i - \lambda_j} \left(\sum_{k=j+1}^i x_k^i \right) \quad \text{for } i > j \quad (5)$$

and $x_j^i = \delta_{ij}$ for $i \leq j$.

However, a closed form for these eigenvectors was only found in the following cases: (a) $b_s = 0$, a model without inactivation; (b) $b_s = b$, with b being a positive constant, a model where each cluster becomes inactivated with the same probability; and (c) $b_s = a s$, now the probability of inactivation of a cluster is proportional to its size and a is the consumption rate of a site.

The case $b_s = 0$ has been already described in the literature [17]. In this case the mass is completely consumed for asymptotic times and it has been found that $n(s, t)$ presents the expected scaling behavior for short times.

We must observe that the main difference concerning fragmentation between this model and the one described by Botet and Ploszajczak [19] is that we consider site consumption instead of bond consumption. They have considered a multiplicative breakup kernel, but when $\beta = 0$ and $\beta = 1$ with $\alpha = 0$ (in their notation), that model is equivalent to ours. Note that in their model, contrary to ours, the mass is constant since it is associated to the sites.

III. MODEL WITH CONSTANT INACTIVATION OF FRAGMENTS

Let us now consider that the inactivation rate of a cluster is constant and independent of its size ($b_s = b$, with $b \geq 0$). All the macroscopic quantities must be either active or inactive (or total). The dynamical equations for the number of fragments are obtained by just adding Eqs. (1)–(3). We obtain that

$$\frac{d}{dt} N_n(t) = a M_n(t) - (2a + b) N_n(t) , \quad (6)$$

$$\frac{d}{dt} N_m(t) = b N_n(t) , \quad (7)$$

$$\frac{d}{dt} N(t) = a M_n(t) - 2a N_n(t) . \quad (8)$$

The equations for the mass are obtained by multiplying Eqs. (1)–(3) by s and then adding them:

$$\frac{d}{dt} M_n(t) = -(a + b) M_n(t) , \quad (9)$$

$$\frac{d}{dt} M_m(t) = b M_n(t) , \quad (10)$$

$$\frac{d}{dt} M(t) = -a M_n(t) . \quad (11)$$

Although these equations can be easily solved for any initial condition $(N_n(0), M_n(0), N_m(0), M_m(0))$, we consider here only the case $M_n(0) = s_0$, $N_n(0) = 1$, and $M_m(0) = N_m(0) = 0$.

The active mass has an exponential decay and the inactive mass grows up to $b s_0 / (a + b)$. Note that the inactive mass diverges in the thermodynamic limit ($s_0 \rightarrow \infty$). The total mass is given by

$$M(t) = \frac{s_0}{a+b} \left(b + a e^{-(a+b)t} \right) . \quad (12)$$

In the beginning of the fragmentation process [$t \ll 1/(a+b)$], the total mass is essentially constant and when t is large it approaches the value of the inactive mass.

The number of active fragments

$$N_n(t) = [1 + s_0(t e^{at} - 1)] e^{-(2a+b)t} \quad (13)$$

increases, reaches a maximum at time t_n and decreases exponentially. Let us consider the thermodynamic limit. The time t_n is given by $t_n = a^{-1} \ln[(2a+b)/(a+b)]$. For short times [$t \ll 1/(2a+b)$], $N_n(t)$ increases linearly in time, in the same fashion as the $b=0$ case. The total number of fragments in the thermodynamic limit

$$N(t) = \frac{abs_0}{(a+b)(2a+b)} + \frac{as_0}{a+b} e^{-(a+b)t} - \frac{2as_0}{2a+b} e^{-(2a+b)t} \quad (14)$$

also has a maximum at time $t_N = 1/a \ln(2)$. This means that the number of fragments reaches a maximum at the same time as the model without inactivation. For short times $N(t) \sim s_0 a t (1 + bt/2)$. If $t \ll 2/b$, the physics is given essentially by the fragmentation process without inactivation and N increases as $N_n(t)$. In this time regime the fragment average size is $\langle s \rangle \sim t^{-1}$. In the limit $t \rightarrow \infty$, $N(t) \sim N_m(t) \sim abs_0 / [(2a+b)(a+b)]$. Note that the number of inactivated fragments always increases in time up to the asymptotic times.

We discuss now the microscopic quantities, the average number of fragments with s sites at time t . All solutions are obtained from Eqs. (1)–(3). The average number of active fragments is given by

$$n(s_0, t) = e^{-(as_0+b)t} , \quad (15)$$

$$n(k, t) = [(s_0 - k - 1)e^{-2at} - 2(s_0 - k)e^{-at} + s_0 - k + 1] e^{-(ka+b)t} \text{ for } k < s_0 . \quad (16)$$

Except for the presence of the inactivation rate b in the time decay, these expressions coincide with the ones for the model without inactivation. For $ka \gg b$, we have a behavior completely controlled by the consumption of sites. In the thermodynamic limit, we have for short times $t \ll 1/a$ the expected scaling form [21,22]

$$n(s, t) \sim t^w s^{-\tau} f(s/t^z) , \quad (17)$$

with $z = -1$, $\tau = 0$, and $w = 2$. On the other hand, for $ka \ll b$, the inactivation drives the decay of the active fragments.

For the average number of inactivated k fragments, we have

$$m(s_0, t) = \frac{b}{as_0 + b} \left(1 - e^{-(as_0+b)t} \right) , \quad (18)$$

when $k = s_0$ and

$$m(k, t) = \frac{b(s_0 - k + 1)}{ak + b} \left(1 - e^{-(ak+b)t} \right) - \frac{2b(s_0 - k)}{a(k+1) + b} \left(1 - e^{-[a(k+1)+b]t} \right) + \frac{b(s_0 - k - 1)}{a(k+2) + b} \left(1 - e^{-[a(k+2)+b]t} \right) \quad (19)$$

if $k < s_0$. Now we consider the limit $s_0 \rightarrow \infty$. Since $m(s_0, t) = 0$ we do not have an ∞ cluster for asymptotic times. On the other hand, we have that $m(k, \infty) \sim s_0$ for all finite k . This means that we have an infinite number of inactivated fragments with finite size for $t \rightarrow \infty$. In fact, for k finite and $t \rightarrow \infty$ we have that

$$m(k, \infty) = \begin{cases} \frac{2bs_0}{ak^3} & \text{for } ka \gg b \\ \frac{2a^2s_0}{b^2} & \text{for } ka \ll b \end{cases} . \quad (20)$$

It is worth mentioning that the k^{-3} behavior is the same as the one found in Ref. [19] for a similar model with bond consumption. The expression for the average total number of fragments is obtained from $n(s, t) + m(s, t)$. In the beginning of the process it behaves like the average number of active fragments and in the end like the number of inactive fragments.

IV. INACTIVATION RATES PROPORTIONAL TO THE FRAGMENT SIZES

We consider here the case of $b_s = as$. Now the probability of a cluster to be inactivated is equal to the probability of a cluster to be fragmented in any two new fragments in a unity time interval. The equations for the macroscopic quantities can be easily obtained from the equations for $n(s, t)$ and $m(s, t)$. However, these equations are difficult to solve. In this model, it is easier to solve first the equations for the microscopic quantities. From Eqs. (1) and (2) we obtain that the average number of active k fragments is given by

$$n(s_0, t) = e^{-2as_0t} , \quad (22)$$

$$n(k, t) = (1 - e^{-2at}) e^{-2akt} \text{ for } k \leq s_0 - 1 . \quad (23)$$

We obtain also the following expressions for the average number of inactivated k fragments:

$$m(s_0, t) = \frac{1}{2} (1 - e^{-2as_0t}) , \quad (24)$$

$$m(k, t) = \frac{1}{2} \left[\frac{1}{k+1} - \left(1 - \frac{k}{k+1} e^{-2at} \right) e^{-2akt} \right]$$

for $k \leq s_0 - 1$. (25)

Let us consider these equations in the thermodynamic limit $s_0 \rightarrow \infty$. We observe that we do not have terms directly proportional to s_0 , implying that $m(k, t)$ for any k is finite during the process. In the limit $t \rightarrow \infty$ we have an infinite cluster or a collection of finite small clusters described by $m(k, \infty) \sim (k+1)^{-1}$ ($k \ll s_0$). This means that either the initial fragment is inactivated in the beginning of the process without the consumption of any site or a site of the initial cluster is consumed. If the latter case happens, two new fragments of very different sizes s_0 (in average) appear and the fragmentation continues. There the inactivation is not so important for the process. Even if a fragment is inactivated, the others maintain the cascade of fragmentation with mass consumption. The k^{-1} behavior is the same found if bond consumption is considered [19].

The mass of the active clusters can be evaluated from the definition $M_n(t) = \sum_k kn(k, t)$. So we have that the macroscopic active quantities are given by

$$N_n(t) = e^{-2at}, \quad (26)$$

$$M_n(t) = \frac{1 - e^{-2as_0t}}{1 - e^{-2at}} e^{-2at}. \quad (27)$$

The number of active fragments is less than 1 because in several samples entering in the average the initial fragment is inactivated [for these samples $N_n(t) = 0$]. So the maximum of the number of active fragments occurs in $t_n = 0$. Near this time, $N_n(t) \sim 1$ and is essentially constant. In the thermodynamic limit the active mass decays initially as $M_n(t) \sim t^{-1}$. This decay comes also from the inactivation of the initial fragment, which implies a considerable loss of active mass in average. For short times ($t \ll 12/a$), $n(s, t)$ has the expected scaling form (17) with $w = 1$, $\tau = 0$, and $z = -1$. The w exponent is in agreement with the scaling relations $N_n(t) \sim t^\phi$ and $M_n(t) \sim t^\epsilon$, with $\phi = w + z(1 - \tau) = 0$ and $\epsilon = w + z(2 - \tau) = -1$. Here the inactivation is responsible for the large mass loss. When the initial fragment is not inactivated, the consumption of sites creates the cascade of small active fragments and we have fragments of all size scales present in the process.

V. INTERMITTENCY ANALYSIS

Our main goal in this section is to study the fluctuations in the number of inactivated fragments $m(s, t)$ of size s in the limit $t \rightarrow \infty$. We could determine the probability of any fluctuation if we knew the probability distribution $P(p_1, p_2, \dots, p_M)$, where p_n is the probability of finding a fragment of size n in the interval $[(n-1)l, nl]$ and $l = s_0/M$ is the size of the bin when we divide the range of possible fragment sizes in M bins. Note that $p_1 + p_2 + \dots + p_M = 1 - p_0$. Here $p_0 < 1$ is a constant probability that no fragment exists at the end of the process.

Unfortunately, we cannot determine $P(p_1, p_2, \dots, p_M)$ in the majority of cases of interest. But we can obtain some information about the distribution

$P(p_1, p_2, \dots, p_M)$ by calculating the moments of the experimental distribution $Q(k_1, k_2, \dots, k_M)$ of fragments at the end of the process, where k_n is the number of fragments in the n th bin. These distributions are related through

$$Q(k_1, k_2, \dots, k_M) = \frac{N!}{k_1! \dots k_M!} \int dp_1 \dots \times \int dp_M P(p_1, p_2, \dots, p_M) p_1^{k_1} \dots p_M^{k_M}, \quad (28)$$

where $N = k_1 + k_2 + \dots + k_M$. In the limit $N \rightarrow \infty$ we have

$$p_n \sim \frac{k_n}{N}. \quad (29)$$

However, in our case this is not true because N is finite and we have to find another way to relate k_m and p_m . This can be accomplished by using the generating function [23]

$$\phi(z_1, z_2, \dots, z_M) = \sum_{N=0}^{\infty} \sum_{k_1, \dots, k_M}' Q(k_1, \dots, k_M) z_1^{k_1} \dots z_M^{k_M} \quad (30)$$

$$= \int dp_1 \dots \int dp_M \frac{P(p_1, \dots, p_M)}{1 - z_1 p_1 - \dots - z_M p_M}. \quad (31)$$

The q th derivatives evaluated at $z_1 = 1, \dots, z_M = 1$ give us the q th moments of the distributions $Q(k_1, k_2, \dots, k_M)$ and $P(p_1, p_2, \dots, p_M)$:

$$\langle k_n(k_n - 1) \dots (k_n - q + 1) \rangle = \frac{q! \langle p_n^q \rangle}{p_0^q}, \quad (32)$$

where the average on the left-hand side is taken over experimental realizations of the process and the one on the right-hand side is taken over the distribution $P(p_1, \dots, p_M)$. This result is equivalent to the one obtained by Bialas and Peschanski [24] for inclusive distributions. Therefore when we calculate the factorial moments

$$F_q = \frac{\langle k_n(k_n - 1) \dots (k_n - q + 1) \rangle}{q! \langle k_n \rangle^q} \quad (33)$$

from the experimental distribution $Q(k_1, \dots, k_M)$ we are actually determining

$$\frac{\langle p_n^q \rangle}{\langle p_n \rangle^q} \quad (34)$$

from the theoretical distribution $P(p_1, \dots, p_M)$.

The behavior of factorial moments (33) can tell us much about the fluctuations of $m(s, t)$. Particularly, if

$$F_q \sim l^{-\phi_q}, \quad (35)$$

it indicates the existence of intermittency in the produc-

tion of inactivated fragments. The scaling behavior in Eq. (35) means the presence of fluctuations in all scales and the absence of any characteristic correlation length.

This behavior is present in the α model [23,24], where the probability of occupation of a bin of size l is given by the probability of occupation of a bin of size $2l$ times a random variable with average value equal to 1, in such a way that the probability of occupation of a bin of size $l = L/2^\nu$ generated at the ν th stage of the cascade process, by successive partitions of initial range L , is a product of ν independent random variables. This gives origin to large fluctuations in the probability of occupation of bins as well to the power law behavior (35).

In order to obtain the experimental realizations over which the factorial moments (33) are averaged, we have simulated the fragmentation-inactivation process with mass loss. In our simulations we have dealt with initial fragment sizes equal to 1024, 2048, and 4096 and with consumption rates a equal to 0.1, 0.5, and 0.9 with $a + b = 1.0$ for both cases $b_s = b$ and $b_s = bs$. We have generated 10^6 samples to calculate the necessary average values.

We have performed the vertical analysis [24,25], i.e., we have calculated the factorial moments for different bins, and the horizontal analysis [23,25], the average of (33) over all bins. Our results are the following. The vertical analysis shows that the factorial moments (33) saturate for small bin sizes (large number of bins), as we can see for the first bin in Fig. 1. Then we can argue that the absence of the power law behavior for the factorial moments F_q indicates that large fluctuations of the extent of the ones in the α model are not present in the dynamics that

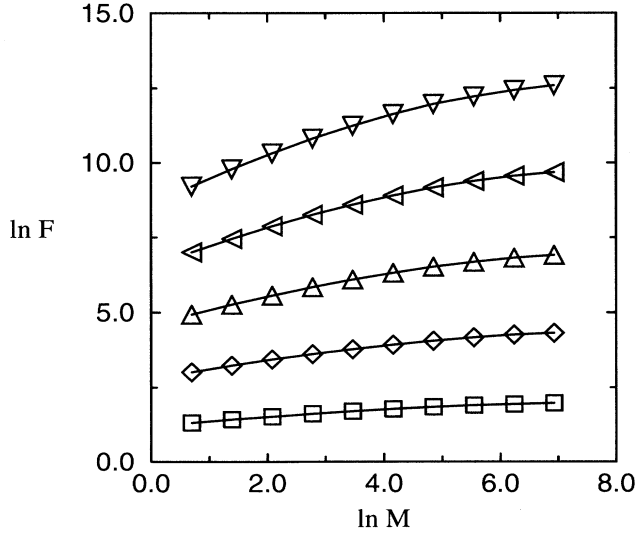


FIG. 1. Log-log plots of the factorial moments $F_q(M)$ versus the number of bins M in the vertical analysis for the first bin with the inactivation rate $b_s = 0.5$ s and an initial fragment size equal to 2048. From the bottom to the top, moments from $q = 2$ to $q = 6$ are shown.

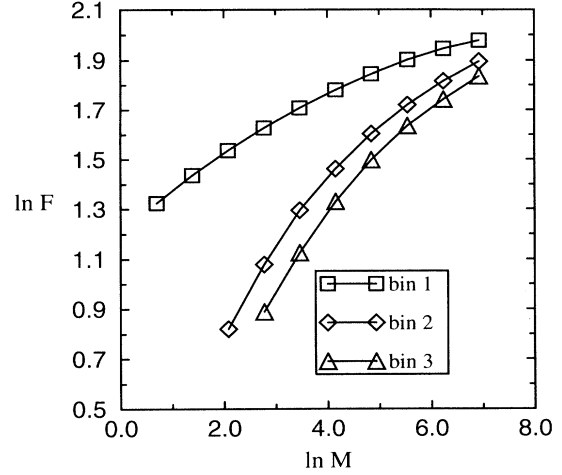


FIG. 2. Log-log plots of the factorial moments $F_2(M)$ versus the number of bins M in the vertical analysis for the three first bins, with the inactivation rate $b_s = 0.5$ s and $s_0 = 2048$.

generates the size distribution of inactivated fragments. This may be confirmed by the fact that contrary to the α model, the bins are not equivalent (see Fig. 2) since the dynamics of the fragmentation-inactivation process favors the production of small fragments, as can be seen from Eqs. (19) and (25) when $t \rightarrow \infty$. In this case, bins that contain small fragments have larger amplitude fluctuations than the bins that contain large fragments; consequently they give higher values to the moments. The horizontal analysis in this case fails completely due to this lack of equivalence between the bins and so does not give the same results as the vertical analysis, as one can see from Fig. 3. The same remarks can be made about the case $b_s = b$ (see Figs. 4 and 5), where the vertical analysis for the first bin and the horizontal analysis are shown.

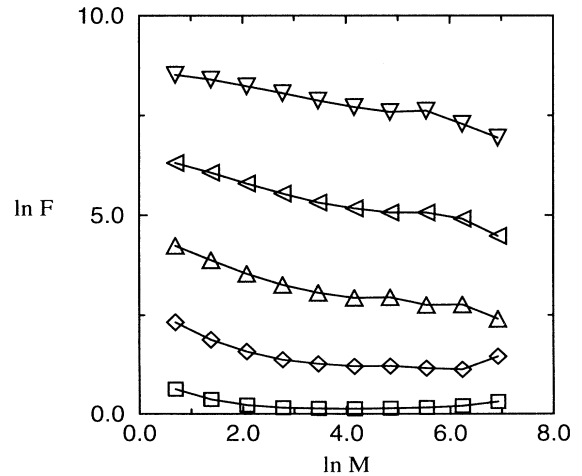


FIG. 3. Log-log plots of the factorial moments $F_q(M)$ versus the number of bins M in the horizontal analysis. Here we have $b_s = 0.5$ s and $s_0 = 2048$. From the bottom to the top, moments from $q = 2$ to $q = 6$ are shown.

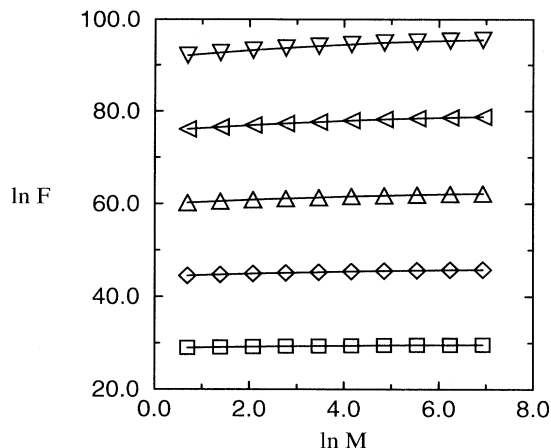


FIG. 4. Log-log plots of the factorial moments $F_q(M)$ versus the number of bins M in the vertical analysis. Here we have $b_s = 0.5$ and $s_0 = 2048$. From the bottom to the top, moments from $q = 2$ to $q = 6$ are shown.

Our results indicate that no intermittent behavior is present in the dynamics of fragmentation-inactivation process, concerning the production of inactivated sites. These results are in contradiction with the ones by Botet and Ploszajczak [19,26] since in these works they have considered a model similar to ours, where bonds are consumed instead of sites, and have performed only a horizontal analysis.

VI. SUMMARY

In this work we have considered several discrete one-dimensional models of fragmentation. The mass of the system decreases by the consumption of the sites and a fragment can be entirely inactivated. The inactivation breaks the cascade of fragmentation randomly. Exact solutions for the average number of fragments $n(s, t)$ have been obtained for the two cases of the inactivation rate b_s of a fragment with s sites: (i) the inactivation rate independent of the fragment size ($b_s = b$) and (ii) the inactivation rate proportional to the fragment size ($b_s =$

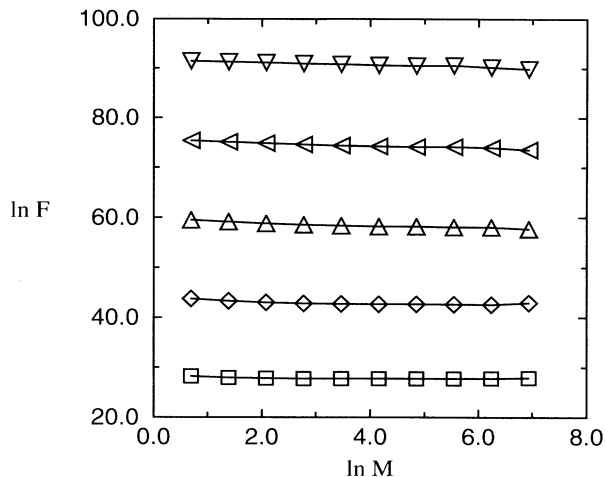


FIG. 5. Log-log plots of the factorial moments $F_q(M)$ versus the number of bins M in the horizontal analysis for the case $b_s = 0.5$. Here the initial size is $s_0 = 2048$. From the bottom to the top, moments from $q = 2$ to $q = 6$ are shown.

as). The scaling behavior of $n(s, t)$ has been discussed for the active and inactive fragments based on analytical results. We studied also the fluctuations in the cluster size distribution for $t \rightarrow \infty$.

When the inactivation rate is independent of the fragment size, the active fragments distribution is the same as the one obtained for the model without inactivation except by a factor $\exp(-bt)$, where b is the constant inactivation rate. If the inactivation rate is proportional to the fragment size, the distributions of active and inactive fragments are very different from the previous case. The inactive fragment distributions at the end of the process do not present intermittency for both cases.

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