

## Criticality in Droplet Fragmentation

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(Received 27 March 1995; revised manuscript received 11 August 1995)

Experiments in rupture of mercury drops have been performed recording the size distribution of the cumulative number of drops for different conditions of rupture. A transition of the distribution from log-normal to scaling behavior is shown. To describe it, a geometric probabilistic model is proposed. Consequently, the rupture can be described as a process of percolation in a Bethe lattice. Then, the rupture process and the transition to scaling can be viewed as a problem of phase transition. Comparison with several fracture experimental data is also made showing the possibilities of this viewpoint.

PACS numbers: 46.30.Nz, 02.50.Cw, 05.70.Fh, 82.20.Wt

The interest in the problem of rupture in liquids is increasing because of the possible application in processes such as combustion as we already reported [1]. In general, the interest in rupture processes is universal and not restricted to liquids as, for example, the process of rock fracture related to log-normal distribution [2]. Some models for fragmentation have been elaborated upon (e.g., [3–6]). Fragmentation has been experimentally studied showing the complexity and the difficulties involved in the study of the behavior of this phenomenon (e.g., [7]). The cumulative number of fragments, i.e., the number of fragments larger than a given size, has special importance in the description of the fracture-rupture processes. This paper is devoted to describing a model able to reproduce the behavior exhibited by the fragment distribution in fracture or rupture as, for instance, that reported by Ishii and Matsushita which measured the size distribution of fragments produced by breaking long thin glass rods [7]. We have performed some experiments on rupture of mercury droplets that give a clear understanding of this model.

The experimental setup is extremely simple. At the height  $h$ , we let fall mercury droplets of about 2.0 mm radius (emerging from a hypodermic needle) directly on a glass Petri dish, where all fragments were collected. Because of the very high surface tension of the mercury (Weber number is of the order of  $10^{-6}$ ), the rupture occurs at the moment of the contact against the dish. Later the fragments were counted and measured with a microscope. The results of the cumulative number of drops versus the drop diameter are plotted in Fig. 1. The solid line represents the theoretical prediction assuming that the distribution is log normal. The distribution obtained shows a clear tendency from a log-normal to a scaled one as the falling height  $h$  is increased. We have performed the Kolmogorov-Smirnov goodness-of-fit test [8] in order to assure this tendency.

For small enough values of the falling height  $h$ , a different characteristic of our results with those of Ishii

and Matsushita [7] is that the log-normal behavior remains in all scales. This is because, in any scale, the shape of our fragments (droplets) is always spherical, whereas in [7], as the rod fragments become of the order of the rod diameter, their shape ceases to be nearly cylindrical and the fracture process evolves, from fracture of nearly one-dimensional objects (cylinders with large aspect ratio) to three-dimensional fracture (objects that already do not resemble cylinders). This change affects the behavior of the fragmentation at small scales, as the authors correctly pointed out.

On the other hand, in our work as in [7], as the falling height is increased, the cumulative number variation starts

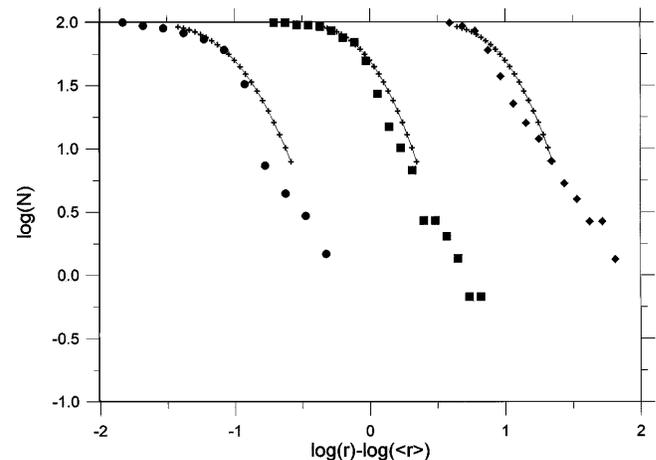


FIG. 1. Experimental results of the cumulative number of droplets (in 10% and log scale) vs  $\log(r) - \log(\langle r \rangle)$ , where  $\langle r \rangle$  is the geometrical mean of the droplet radii. The falling heights are 0.5 m (circles), 1 m (squares), and 5 m (diamonds). Note that, to clarify the plot, we have shifted one unit to the left of the results for  $h = 0.5$  m and one unit to the right of the results for  $h = 5$  m. Solid lines represent the theoretical prediction (related to the error function) assuming log-normal distributions. Transition to a scaling law when falling height is increased is evident.

to show a power law dependence on their size, especially for the larger droplets. The viewpoint presented in [4] gives a nice geometrical description of the power law distribution but the transition from log normal to scaling at our present knowledge has not yet been described.

We propose a geometrical model which does not take into account the physical mechanism of rupture and, as in [4], we use essentially the same procedure but conditioned by a parameter  $p$  which represents the probability of rupture of a given drop. So let us start with  $N_0$  initial droplets of unit radius and break one of them in  $F$  fragments (smaller droplets) with probability  $p$ . Thus  $pN_0$  droplets will break giving  $FpN_0$  fragments of radius  $r_1 = F^{-1/3}$ , and  $(1-p)N_0$  droplets will stay without breaking. Then, as a consequence of the first breakage, we have a collection of  $(1-p)N_0$  first generation droplets with unit radius and  $FpN_0$  smaller droplets with radius  $r_1 = F^{-1/3}$  (droplets of second generation).

Now let us apply the same procedure to these smaller droplets, i.e., a fraction  $p^2FN_0$  of droplets will break to even smaller droplets. As each one will break into  $F$  pieces again, the number of even smaller droplets (droplets of third generation) will be  $p^2F^2N_0$ . The new collection of droplets at this step of the procedure will be  $(1-p)N_0$  drops of radius  $r = r_0 = 1$ ,  $(1-p)pFN_0$  droplets of radius  $r = r_1 = F^{-1/3}$ , and  $p^2F^2N_0$  droplets of radius  $r = r_2 = F^{-2/3}$ . The process can easily be continued and the cumulative number can be expressed in the  $k$ th generation for  $k \gg 1$  as

$$N = (1-p)N_0(1 + pF + p^2F^2 + \dots + p^{k-1}F^{k-1}) \\ \sim p^k F^k,$$

where  $1 + pF + p^2F^2 + \dots + p^{k-1}F^{k-1}$  is the sum of a geometric progression with a common ratio  $pF$ . In order to not have converging series, the common ratio  $pF$  must be larger than 1, so

$$p > p_c = 1/F. \quad (1)$$

On the other hand, if this distribution is scaled (i.e.,  $N \sim r^{-x}$ ), we can obtain for  $k \gg 1$  the following expression for  $x$ ,

$$x = 3 \left( 1 + \frac{\ln p}{\ln F} \right).$$

For  $x$  to be positive the condition  $p > p_c = 1/F$  must be accomplished, which is the same condition obtained from the nonconvergence of the sum of the geometric progression.

This gives us some kind of critical value or threshold above which this model can show scaled distributions. Otherwise, if  $p < p_c$ , the breaking process will stop in some stage of the rupture chain and the cumulative number, in this last case, will saturate in small scales. As we will see, this behavior resembles the corresponding one to the log-normal distribution law.

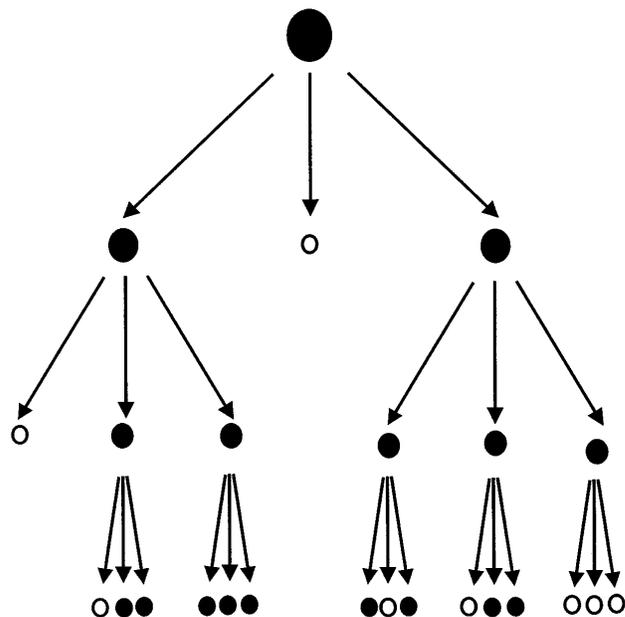


FIG. 2. A drawing of the breaking process in this probabilistic geometric model. Full circles indicate that fragmentation occurs, empty circles indicate that breaking chain ends in that fragment. This is indeed the characteristic feature of a Bethe lattice.

In Fig. 2, we illustrate with a drawing the process already described. One of the initial drops gives rise to three new droplets. Only two of them undergo the process of subsequent division, each generating three new fragments. The other drop does not continue the process. In this way we can understand this scheme as a lattice through which a process of percolation occurs. The system will generate a cluster whose length will be finite for small  $p$  (i.e.,  $p < p_c$ ) and infinite or very long (i.e., percolation) if  $p > p_c$ . Besides, we can see that this cluster is the reproduction of a Bethe lattice, in which the critical probability for percolation to occur is precisely  $p_c = 1/3$ .

The description of the rupture process in this way has been translated to the problem of percolation in the Bethe lattice. Thus we may try to calculate the influence of  $p$  in the percolation length  $\xi$ .

As percolation must occur above  $p = p_c$ , then we may formulate that

$$\xi \sim \frac{1}{(p_c - p)^\nu}. \quad (2)$$

Following the standard procedure of renormalization, let us set that in the lattice a cluster is occupied when at least one of the bonds exists. As an example, in Fig. 2 the first cluster is, of course, occupied, because the first drop was broken and the two fragments of the extremities also continue the process. The third step shows an empty cluster (the upper one) and four occupied

clusters. In general, when each node of the Bethe lattice has  $F$  terminals, the above definition of occupied clusters applies. In such a way the probability of occupation of a bond is the probability of breaking the first drop ( $p$ ) and also any of the subsequent fragments, i.e.,

$$\begin{aligned}
 p' &= p^{F+1} + (F + 1)p^F(1 - p) \\
 &+ \binom{F + 1}{2} p^{F-1}(1 - p)^2 + \dots \\
 &+ \binom{F + 1}{F - 1} p^2(1 - p)^{F-1}, \quad (3)
 \end{aligned}$$

where this sum comprises up to the probability that  $F - 1$  of the *new* drops do not break whereas the *mother* drop and one of their *daughters* was broken [last term in Eq. (3)].

The first term on the right hand side of (3) describes the probability for breakage of all the drops, the second is the probability of breakage of all except one drop, and so on.

$$\begin{pmatrix} a \\ b \end{pmatrix}$$

represents combinations of  $a$  objects in groups of  $b$ , where  $b < a$ . Really, in this formulation is hidden the possibility that one of the nonbroken drops is the *mother*, but it is to be expected that, near the percolation threshold, this process would not be significant.

By the binomial formula, (5) can be transformed to

$$p' = 1 - \binom{F + 1}{F} p(1 - p)^F - (1 - p)^{F+1}. \quad (4)$$

Now we are able to rescale (coarse graining)  $r \rightarrow F^{1/3}r$ . After this rescaling, we will have a new percolation length  $\xi' = \xi/F^{1/3}$ . The standard renormalization procedure leads us to the following result:

$$\nu = \frac{1}{3} \frac{\ln F}{\ln(F + 1) + (F - 1)\ln(1 - 1/F)}, \quad (5)$$

from where we obtain the values  $\nu_{\max} = 0.569837$  as the maximum value of the exponent corresponding to two fragments and  $\nu_{\min} = 1/3$  corresponding to a large  $F$ .

It is not difficult to make a simulation of this percolation process obtaining for each value of  $p$  the value of the percolation length represented with dots in Fig. 3 and the analytic behavior of Eq. (2) for both extreme values of  $\nu$ . As can be seen there is a nice similarity in behavior. In Fig. 4 we show the correspondence of the cumulative number of fragments predicted by this model with the experimental data. The simulation was performed for  $F = 2$  and  $p = 0.82$ . In Fig. 5, the variation of the cumulative number of fragment distribution for different values of  $p$  according to this model is shown.

This proposed viewpoint indicates that rupture behaves as a critical phenomenon, at least in this description. The model presented here describes the process of transition experienced by the distribution of fragments in breaking

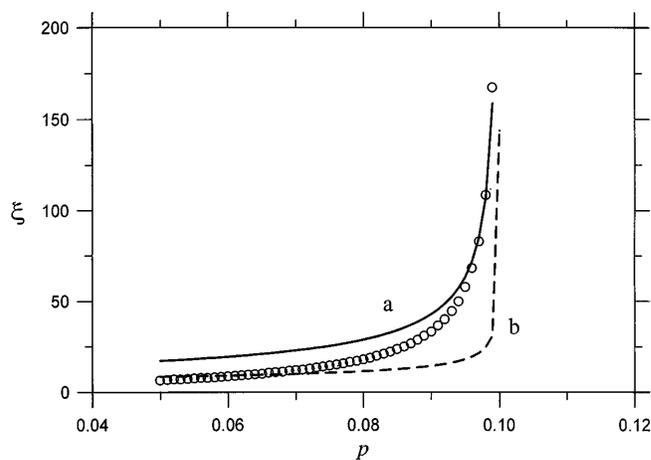


FIG. 3. Variation of the penetration length in the Bethe lattice as a function of  $p$  for  $F = 10$ . Circles indicate the behavior according to the proposed model. Curves  $a$  and  $b$  indicate the variation of  $\xi$  with  $p$  according to Eq. (2) for  $\nu = \nu_{\max} = 0.569837$  and  $\nu = \nu_{\min} = 1/3$ , respectively.

phenomena. Although this simple model has a fixed  $p$ , it reproduces very well the behavior of the cumulative number of droplets in the rupture process. We point out how universal properties can be obtained from this oversimplified model. We do not try to reproduce with this model the actual dynamics of the droplet rupture, which is, surely, much more complex than our model, even with a variable value of the parameter  $p$ , depending on the  $k$  step. However, we have simulated the rupture process with  $p$  variable, and we have obtained the same qualitative results.

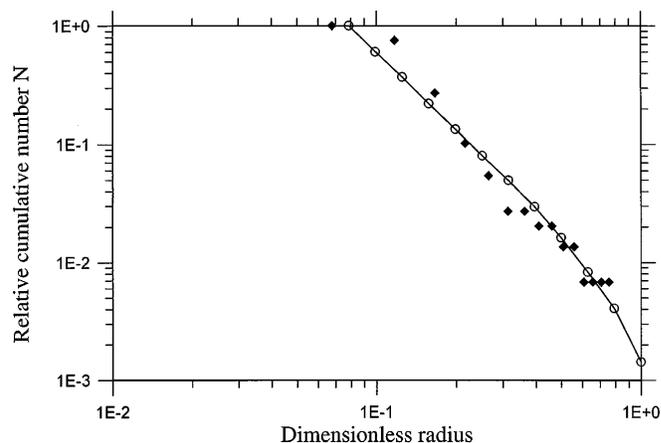


FIG. 4. Similarity in behavior of the model with an experimental realization. The comparison was, in this case, performed with data (diamonds) corresponding to our experiment for  $h = 1$  m, and the theoretical line (full line with empty circles) for  $F = 2$  and  $p = 0.82$ . Other curves can also be fitted by using this model with an adequate selection of  $F$  and  $p$ .

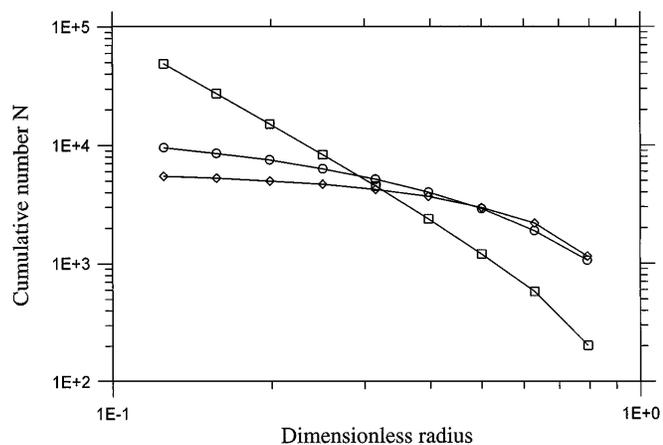


FIG. 5. Variation of cumulative number  $N$  vs dimensionless radius, according to this model. We point out that, for different  $p$ 's, the shape of the curves behaves as the experimental results for different falling heights. Squares represent  $p = 0.9$ , diamonds  $p = 0.4$ , and circles  $p = p_c = 0.5$ , with  $F = 2$ .

This work was carried out while one of the authors (O.S.) was visiting the U.N.E.D. in Madrid, Spain. The financial support by the Vicerrectorado de Centros Asociados of the U.N.E.D. is gratefully acknowledged. This

work was partially supported by the Dirección General de Investigación Científica y Técnica (DGICYT, Spanish Ministry of Education and Science) under Project No. PB91-0221. We also want to thank Professor Liñan, Professor Castillo, and Professor García-Ybarra for fruitful discussions.

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