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DROPLET SIZE DISTRIBUTION IN A PERCOLATION MODEL FOR EXPLOSIVE LIQUID DISPERSAL

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A very complicated problem on irregular motion is involved in theoretical analysis of liquid deformation during dispersal and explosive break-up, which in general cannot be solved accurately. The chaotic (explosive) dispersal of a liquid is here related to the break-up in an infinite cluster as studied in percolation theory. The drop size distribution is derived theoretically. If the dispersal is planar, the standard empirical relations are obtained (the Rozin-Rammler law and Weibull distribution), but in the three-dimensional case, there are deviations from them. Measurements have also been made on dispersal for a concentrated elastoviscous liquid based on a polymer on wire explosion in a cylindrical volume. The measurements on the whole agree satisfactorily with the theory.

1. The following liquids are examined here: Newtonian ones (in particular, ideal ones) with surface tension and polymer ones, which have internal entropy elasticity [1]. For a sufficiently concentrated polymer liquid ($\geq 1\%$), the surface tension is usually unimportant, since virtually always $G a_0 / \alpha \gg 1$, in which G is the elastic modulus, α the surface tension, and a_0 the minimum characteristic dimension, which is discussed in detail below.

At $t = 0$, a bounded liquid volume with characteristic dimension R_0 acquires kinetic energy E_0 due for example to an explosion at the center. This concerns particularly the electrical explosion of a wire or a detonation within a bounded volume (see [2-4] and Sec. 3 below). In such cases, there are several factors that lead almost instantly to irregular chaotic deformation, which precedes the break-up and favors the latter. One of them is the shape imperfection or inhomogeneity in the exploding wire or detonator, which leads to initial irregularity in the velocity pattern. Another is that the explosion-cavity expansion is accompanied by Rayleigh-Taylor instability [5-7], which is the first stage in the irregular motion. As that form of instability develops, the motion becomes more complicated and chaotic, and in the nonlinear stage of perturbation growth, vortices arise at the tips of the fingers. To some extent, the break-up itself indicates that there are irregular motions, and accentuation of the chaotic motion is evident at the stage where there are separate droplets, which is evidently due to new modes occurring, particularly on expansion in the vacuum, which can occur in the motion of the continuous volume at least as small perturbations.

Sometimes, one expects that the kinetic energy in the irregular motions arising from a central explosion will be $E \sim E_0$; this is evident from estimates of the dissipative losses. Also, in general refining E does not affect the theory and merely has a quantitative effect on the results.

We assume that the central explosion almost immediately gives rise to internal deformations corresponding to many degrees of freedom, which absorb much of E_0 . Such motion has been discussed [8] and is due either to turbulence or to initial inhomogeneity in the velocity pattern from the explosion.

In general, the dispersal and explosive break-up at present do not allow of a formal discussion of the internal irregular motion excitation. Also, there are some examples where

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there is virtually no excitation of the internal degrees of freedom. However, in some cases such as the ones above, it is a natural assumption that almost instantly one gets inertial motion having $E \sim E_0$, and because many degrees of freedom are excited, that motion rapidly becomes irregular, isotropic, and chaotic. Such cases are considered here.

The isotropic chaotic motion leads to drops becoming detached from the outer surface of the original continuous volume; within it, disruption results, which produces new areas of free surface and isolated droplets. That picture includes the observed [2-4] mass formation of free surfaces in growing cavities during explosive dispersal for a cylindrical water volume. An important feature used subsequently is that in such isotropic motion, one cannot get droplets smaller than a certain size governed by the physical mechanism at the microscopic level and by the characteristics of the liquid. This can be demonstrated for some important cases.

In isotropic random deformation, each individual droplet has a deformation rate $\dot{\gamma}$ such that

$$(1/2)\rho R_0^3(\dot{\gamma}R_0)^2 \sim E, \quad \dot{\gamma} \sim (E/\rho R_0^3)^{1/2}, \quad (1.1)$$

in which ρ is density.

In a strong explosion, the dissipation associated with internal friction can be neglected for a time usually sufficient for the break-up to go almost to completion. When however dissipative processes predominate, the break-up can evidently be only regular.

In a newtonian liquid, the kinetic energy of an individual droplet with characteristic size a must at least exceed the surface energy:

$$(1/2)\rho a^3(\dot{\gamma}a)^2 \geq \alpha a^2. \quad (1.2)$$

Equality in (1.2) corresponds to the size of the smallest drop a_0 capable of becoming detached from the bulk, so from (1.1)

$$a_0 \sim \left(\frac{\alpha R_0^5}{E}\right)^{1/3} \sim \left(\frac{\alpha R_0^2}{\rho E_*}\right)^{1/3} \quad (1.3)$$

in which E_* is the specific kinetic energy in the deformation.

Turbulent deformation involves the internal scale in the turbulence ℓ_0 in no way restricting a_0 , since at distances a such that $a_0 < a < \ell_0$, the differences in velocity may be quite sufficient to continue the droplet break-up for $a < \ell_0$ [9].

In a concentrated polymer liquid, the kinetic energy in the deformation required to detach a droplet must at least exceed the elastic energy related to the entropy elasticity in the macromolecules:

$$(1/2)\rho a^3(\dot{\gamma}a)^2 \geq G a^3.$$

On the assumption that the relative extension in the disruption of a polymer liquid $\lambda_* = O(1)$, that inequality can be considered as the condition for the dynamic head $(1/2)\rho \cdot (\dot{\gamma}a)^2$ exceeding the failure stress $\sigma_* \sim G\lambda_*^2 \sim G$. It is also assumed that one can neglect the viscous dissipation and the elastic-stress relaxation in a strong explosion during the ongoing dispersal, which is evident from estimates. Then for a polymer liquid

$$a_0 \sim \left(\frac{GR_0^5}{E}\right)^{1/2} \sim R_0 \left(\frac{G}{\rho E_*}\right)^{1/2}. \quad (1.4)$$

At the start, when the deformation has not had time to become isotropically chaotic, some parts of the liquid may have deformation rates exceeding the $\dot{\gamma}$ from (1.1), which in principle may result in a certain number of droplets with size less than a_0 , which distorts the picture somewhat.

Above, we have examined liquid break-up under vacuum. If there is surrounding air and there are high velocities in the relative motion, one almost instantly gets a chaotic deformation with kinetic energy $E \sim \rho_1 U_0^2 R_0^3$, and thus explosive dispersal (ρ_1 is the density of air and U_0 is the speed in the relative motion between the center of mass of the liquid and the air). The above formulas still apply, in particular (1.3) and (1.4).

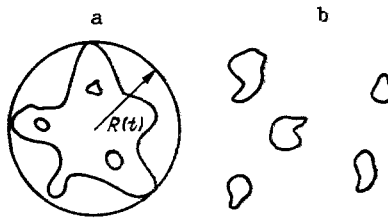


Fig. 1

2. We now consider the size distribution. From Sec. 1, we consider the entire volume as a cluster (or clusters after the start of break-up) composed of individual droplets P with size a_0 . It is assumed that the entire volume, and any droplet detached from it, will consist of clusters of size a_0 , so the sizes of the detaching droplets may exceed a_0 , and that fairly substantially. An important new feature is that the positions of the individual droplets P are random, which assumption is justified for rapid randomization (by comparison with the duration of the break-up) as a result of the motion initiated by the central explosion, which usually occurs for large E, when the Reynolds number is $Re = (\rho E / R_0 \mu^2)^{1/2} \gg 1$ (μ is the effective viscosity). Correspondingly, at time t the droplets P are randomly distributed in a region having characteristic dimension $R(t) \sim R_0(1 + \dot{\gamma}t)$. Initially, the expanding medium is compact (Fig. 1a), but then it splits up into drops (clusters), which in turn break up further (Fig. 1b). We assume that the space is split up into cells with size a_0 , and with probability

$$p(t) = \frac{R_0^d}{R_0^d(1 + \dot{\gamma}t)^d} = (1 + \dot{\gamma}t)^{-d} \quad (2.1)$$

a cell will be occupied by a droplet P, while with probability $1 - p$ it will be free (d represents the dimensions of the space characterizing the break-up). For $a_0 \ll R_0$, the Fig. 1 picture is that of break-up in an infinite cluster composed of filled cells. The break-up or formation of an infinite cluster has been examined in percolation theory [10].

As $p(t)$ decreases during the expansion, gaps appear in the infinite cluster (the original connected liquid volume), and at time t_* , when $p(t_*) = p_*$ (critical value), the infinite cluster is destroyed. The body is converted to a group of finite clusters (drops), which may in turn break up. The nodal treatment (cells with face length a_0) gives $p_* = 1$ ($d = 1$), $p_* = 0.593$ ($d = 2$, square lattice) or $p_* = 0.311$ ($d = 3$, simple cubic lattice) [10], so t_* can be calculated from (1.1) and (2.1).

To derive the size distributions, we used results from percolation theory. The probability of a finite cluster existing, composed of s nodes w_s for $p < p_*$ ($t > t_*$), is normalized in such a way that $\sum_{s=1}^{\infty} w_s = 1$ and for sufficiently large s has the asymptote

$$w_s = w(s) = Ks^{-\theta+1} \exp(-bs^\zeta), \quad (2.2)$$

in which K is a normalization factor and $b = b(p, d)$; $\theta = \theta(\bar{p}, d)$:

$$\theta(0 < p < p_*) = \begin{cases} 0, & d = 1, \\ 1, & d = 2, \\ 1, 5, & d = 3, \end{cases} \quad \theta(p_*) = \tau = \begin{cases} 2, & d = 1, \\ 2, 0, & d = 2, \\ 2, 1, & d = 3, \end{cases} \quad (2.3)$$

while $\zeta = 1$ for $0 < p < p_*$ and $\zeta = 0$ for $p = p_*$. The [10, 11] results were used to estimate b: $b(p = 0.15) \approx 0.693$, $d = 2$; $b(p = 0.15) \approx 0.36$, $d = 3$; $b(p_*) = 0$, $d = 2$ and 3.

We note that $w_s = V_s/V_0$, in which V_s is the total volume of all clusters composed of s nodes, with V_0 the total volume of the medium.

With accuracy sufficient for the subsequent purposes, (2.2) can be used with $s = O(1)$ because correspondingly the s dependence of $\ln w_s$ incorporates not only the power-law term but also the logarithmic one. The scope for extrapolating (2.2) to $s = O(1)$ is indicated also by the comparison below between the frequency curve derived from that extrapolation and empirical data [12], so we used (2.2) to replace the sign of asymptotic equality everywhere by the usual equal sign.

We introduce the drop volume $V = (4/3)\pi a_0^3 s$. Equation (2.2) gives $w(s)ds = f(V)dV$, in which $f(v)$ is the probability density that the drop volume falls in the range from V to $V + dV$, and correspondingly

$$f(V) = \frac{3K}{4\pi a_0^3} \left(\frac{3V}{4\pi a_0^3} \right)^{-\theta+1} \exp \left[-b \left(\frac{3V}{4\pi a_0^3} \right)^\zeta \right]. \quad (2.4)$$

Any dynamic factor showing random and isotropic behavior can only affect a_0 but not the form of the universal (2.4), which is of geometrical origin.

The proportion of the volume accounted for by drops with volumes greater than V is $R = \int_V^\infty f(V)dV$, and from (2.3) is

$$R(V) = \exp(-z) \text{ for } d=2, \quad R(V) = 1 - \frac{\Gamma_z(1/2)}{\Gamma(1/2)}, \text{ for } d=3, \quad (2.5)$$

$$z = 3Vb/(4\pi a_0^3)$$

where $K = b$ from the normalization condition for $d = 2$ and $K = b^{1/2}/\Gamma(1/2)$ for $d = 3$, while $\Gamma(\cdot)$ and $\Gamma_z(\cdot)$ are the gamma function and the incomplete gamma function.

As $V = (4/3)\pi x^3$, in which x is characteristic droplet size, the first formula in (2.5) takes the form of the empirical Rozin-Rammler law [13, 14]:

$$R(x) = \exp[-b(x/a_0)^3], \quad (2.6)$$

and $W(x) = 1 - R(x)$ gives a Weibull distribution [14]. The dW/dx frequency curve defined by (2.6) corresponds to empirical approximations used for spraying and the dispersal of liquids by spray jets [12], which indirectly confirms that (2.2) can be extrapolated to $s = 0(1)$.

The [15] measurements also relate to spraying from jets and lead to $R(x) = \exp[-(x/x_m)^n]$, in which $n = 3.21$, while results from other sources mentioned in [15] give $n = 2-4$ (x_m is the characteristic drop size), which agrees satisfactorily with (2.6). Out of the more detailed differential characteristics corresponding to the Rozin-Rammler law, the surface and bulk distributions agree satisfactorily with the [15] results; there is however a discrepancy with the numerical distribution, which may correspond to the (2.5) conclusion that when the break-up is distinctly of volume type ($d = 3$), there should be deviations from the Rozin-Rammler law, which should be seen primarily in the detailed differential characteristics.

For $d = 2$, the Rozin-Rammler law and the Weibull distribution are derived here theoretically for liquids, while the parameters appearing in them have been calculated. For $d = 3$, there are deviations from that law and distribution.

In (2.6), the radius x corresponds to a drop of approximately spherical shape. When optical measurements are made on the sizes of drops breaking up in flight, which have complicated shapes, one can identify the effective radius of inertia, which satisfies [16] $x \sim a_0 s^{1/2}$ ($0 < p < p_*$, $d = 3$). If such an effective characteristic is measured, one has to compare theory with experiment on the basis that $s \sim (x/a_0)^2$, $V \sim a_0^3 s \sim a_0 x^2$, $z \sim V/a_0^3 \sim (x/a_0)^2$ so the exponent 3 changes to 2 in (2.6).

The mean drop volume is proportional to a_0^3 , while the mean number is $N \sim (R_0/a_0)^3$. Then (1.3) and (1.4) give for newtonian and concentrated polymer liquids that

$$N \sim \rho E_* R_0 / \alpha, \quad N \sim (\rho E_* / G)^{3/2}.$$

These universal results correspond to the limit of isotropic chaotic deformation. The other limit is regular break-up and corresponds to capillary and bending break-up in jets, the break-up of edge ridges in free films, Rayleigh-Taylor instability in liquid films accelerated by gas-pressure differences [5-7, 17-22], etc.

3. We have made measurements on explosive break-up for a 3% solution of polyethylene oxide type PEO-FPR (molecular mass $4 \cdot 10^6$ g/mole), which is a typical elastoviscous liquid [23]. The apparatus was similar to that used in [2-4]. The solution was contained in a cylindrical paper tube with radius 2 cm and length 15 cm placed vertically between two sleeves. A tungsten wire ran along the axis, whose ends were attached to the sleeves. The electrical explosion dispersed the solution mainly in the radial direction, which provided close to planar ($d = 2$) break-up (in the plane of section of the originally cylindrical volume).

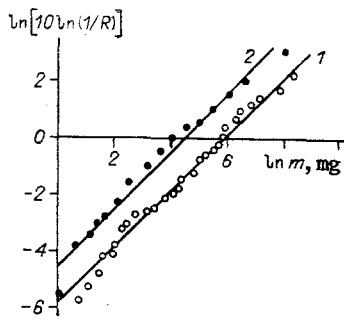


Fig. 2

The droplet distribution was recorded with shields covered by millimeter paper at 1.5 m from the point of explosion. We used up to 19 shields, with the size of the millimeter paper on each 20 cm wide and 1 m long. This recorded up to 40% of the drops. The liquid was at 10°C at the time of explosion and the air temperature was -20°C. The drops reaching the paper froze rapidly, which largely eliminated the evaporation and the resulting error in determining the mass, while also facilitating weighing.

For weighing, the frozen drops were cut out together with the millimeter paper (the paper area somewhat exceeded the area of the attached drop). The weight of the drop and paper was recorded together with the area of the paper and the area of the base of the drop. The drop weight was determined by subtracting the known weight of the paper. This was done for all drops. The weighing with an analytical balance was to ~0.1 mg.

For comparison with experiment, we transform (2.5) to

$$R(m) = \exp\left(-\frac{m}{m_0}\right) \text{ for } d = 2, \quad R(m) = 1 - \frac{\Gamma_z(1/2)}{\Gamma(1/2)} \text{ for } d = 3, \quad z = \frac{m}{m_0} \quad (3.1)$$

($m = \rho V$ is mass, $m_0 = (4/3)\pi a_0^3 \rho/b$).

For $d = 2$, the first formula in (3.1) gives

$$\ln(10 \ln 1/R) = \ln 10 + \ln m - \ln m_0. \quad (3.2)$$

Figure 2 compares the (3.2) theoretical result with the measurements for the PEO-FPR; the circles denote measurements for an explosion with specific energy E_* 27.7 J/g, and the filled circles are for E_* 40 J/g. Lines 1 and 2 show theoretical (3.2), with $m_0 = 3650$ mg for 27.7 J/g and 995 mg for 40 J/g. The measurements fit the straight lines with slopes of 1, which confirms (2.5), (3.1), and (3.2), which predict $\ln(10 \ln R^{-1}) \sim \ln m$ for $d = 2$.

In accordance with (1.4), one should have $m_0 \sim a_0^3 \sim E_*^{-3/2}$. The measurements gave $m_{01}/m_{02} = 3650/995 = 3.67$, which differs from the theoretical $(40/27.7)^{3/2} = 1.74$, possibly because much of the liquid evaporates at the higher specific energy, which additionally reduces m_{02} .

In these experiments, the break-up was close to planar. Interest attaches to the break-up of spherical volumes of polymer liquids on central explosion, as this would test the second (3.1) formula.

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