Editors' Suggestion

Scaling Laws of an Exploding Liquid Column under an Intense Ultrashort X-Ray Pulse

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A general formulation of the partial destruction of a liquid object in vacuum after the sudden deposition of a very large amount of energy is proposed. That energy instantaneously raises the pressure of a portion of the liquid to extreme values and changes its state, which causes its explosive expansion into vacuum and against the rest of the liquid object. When the deformable object is a liquid capillary column, the model reduces to a universal equation for the evolution of the expanding gap between the two sides of the exploding liquid column. The theoretical analysis contemplates two asymptotic stages for small and large times from the initiation of the blast, whose asymptotic solutions are fitted to available experimental data. A universal approximate analytical solution is obtained. A complete dimensional analysis of the problem and an optimal collapse of experimental data reveal that the proposed solution is in remarkable agreement with experiments of a jet exploding after being irradiated by an ultrashort and intense x-ray pulse from an x-ray free electron laser.

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The interaction of high energies with matter is a subject of fundamental importance in applied physics [1,2]. There is ample research literature on the reaction of condensed matter to the localized deposition of a very large energy density by diverse means (e.g., by large electric shocks, laser and ion beams, etc.), especially in the area of inertial confinement fusion. Extensive related research on the hydrodynamic processes due to the very fast local vaporization, like shock-driven hydrodynamics, liquid compressibility phenomena, bubble implosion, etc., appears in the literature for spherical [3,4] and cylindrical geometries [5,6]. In these latter cases, the energy fluxes ranged from 10^{10} to 10^{12} W/cm^2. The advent of powerful energy sources like the free electron lasers (XFELs) has raised the energy fluxes above 10^{22} W/cm^2, with the top $>10^{23}$ W/cm^2 mark announced by the project ELI-NP at Magurele [7], the largest known so far in our planet. The local ultrafast (femto- and attosecond-scale) deposition of these extreme energy densities have been crucial to observe and test new phenomena and more diverse geometries [8], overcoming the effects of the energy release history of previous slower deposition means [5,6].

The physics of intense blasts against deformable (liquid) objects is of fundamental importance in areas as varied as nanosurgery [9], serial femtosecond crystallography (SFX) [10], the study of extreme physical properties and strange phases of matter [11,12], or testing the equations of state of matter from cosmology [13,14] to the processing or production of new materials [15]. In general, the extremely large local pressures appearing in the liquid in very short times and the need to quantify them are crucial objectives of research. When the liquid is a closed object with free surfaces (e.g., a sphere or a rod [8]), the scaling laws of those pressures, the input energy fraction going to pressure, the evolution of the blast, or the blast shape factor against the liquid being destructed need to be determined to predict the effects. In SFX, the damage caused to the samples upstream of the blast is the subject of increasing attention [16].

In this work we focus on the destruction of a cylindrical rod of liquid (e.g., a capillary jet) after the rapid local deposition of a very large energy density [8] in processes like the analysis or processing of materials with synchrotron, SFX, etc. This deposition causes an explosion that expands partially in vacuum and partially against the liquid rod in the axial direction, violently dividing the cylinder into two sections. Unlike the radial actions that occur in exploding wires, causing strong radial compression phenomena only [5], the explosions here studied imply a geometrical degree of freedom in the axial direction that is absent in the former. This implies the appearance of a variety of new physical phenomena [8]. The essential difference with other processes where a liquid cylinder breaks up is the extreme value of the power locally supplied: it can be trillions times larger than that of natural mechanical methods (e.g., capillary breakup, or the sudden jet obstruction or its transverse disruption with a projectile). Here we propose a general formulation of the problem, applying it to a cylindrical liquid column. The physics (hydrodynamics) of the blast is described obtaining the expansion velocities, the peak compression stresses undergone by the liquid column, and the energies involved.

General compact formulation of blasts in partial contact with deformable objects.— Consider an amount of condensed deformable matter M surrounded by vacuum

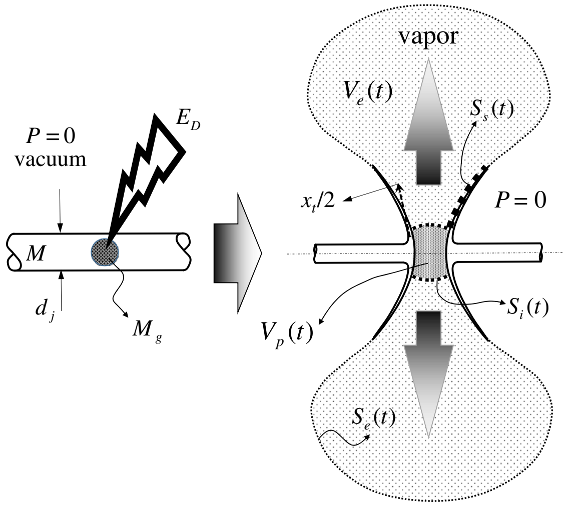

FIG. 1. General sketch of the problem. Thin, medium, and thick dotted lines indicate $S_e(t)$, $S_i(t)$, and $S_s(t)$, respectively. $S_p(t)$ is the surface of $V_p(t)$ in contact with the liquid. The instantaneous radial velocity $x_t/2$ of the expanding lamella is indicated.

(Fig. 1). In this work, that matter is in the form of a cylindrical rod. An amount of energy E_D exceeding by orders of magnitude that to vaporize a portion M_g of that matter is suddenly injected in it. As a consequence, it violently expands as a blast both into vacuum and against M (Fig. 1). The expanding hot gas domain $V_g(t)$ can be consider to comprise two virtual subdomains $V_p(t)$ and $V_e(t)$ (from now on *pushing* and *expanding* volumes, respectively) separated by a fluid surface $S_i(t)$, as formally defined in the Supplemental Material [17]: (i) $V_p(t)$ pushes and deforms M by dominant pressure forces, (ii) the mass and energy of $V_e(t)$ are constant by definition while it expands into vacuum, and (iii) $V_e(t)$ is assumed charge neutral. By virtue of two latter conditions, $V_e(t)$ does not make any work on M. Those definitions do not impose any artificial restriction on the natural evolution of the total gas domain $V_g(t)$, but they provide a drastic simplification, as follows. Defining $V_o = V_p(t_0) + V_e(t_0)$ as the initial energized volume at the initial instant t_0, the volume fraction $\chi = V_p(t_0)/V_o$ is a fixed problem parameter. Thus, given that the analysis of V_e is irrelevant since its energy is constant and its evolution is decoupled from M, one can write the following compact equation of conservation of energy that governs the coupled evolution of M and V_p:

$$\int_{t_0}^{t}\int_{S_p+S_i} P\mathbf{v}\cdot\mathbf{n}\,dAdt' + \frac{P_o V_p(t)^{1-\gamma}(\chi V_o)^{\gamma}}{(\gamma-1)} = \chi E_D, \qquad (1)$$

where the first term in the left side is the *total* work made by V_p on M through the fluid surfaces S_i and S_p (the surface of V_p in contact with M; see Fig. 2) since the beginning of the blast ($\mathbf{v}$ and $\mathbf{n}$ are the velocity and unit normal vectors of the surfaces), and the second is the internal energy left in V_p at a given t. The initial conditions are $V_p(t_0) = \chi V_o$, $P(t_0) = P_o$, and $P_o V_o/(\gamma-1) = E_D$. Since the pressure is initially the same for V_p and V_e, χ is also the injected energy fraction contained in V_p at the beginning of the

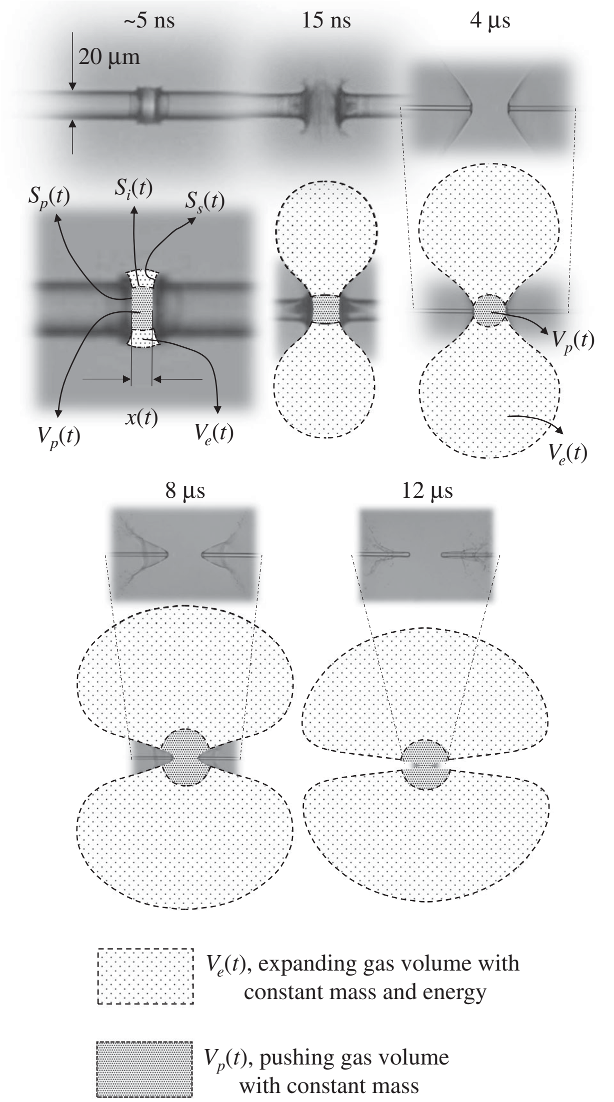

FIG. 2. Evolution of the blast induced by a strong x-ray laser pulse (photon energy 8.2 KeV, 0.75 mJ, duration 30 fs) on a liquid microjet of 20 μm discharged in vacuum [8]. The effective beam diameter is approximately 1 μm. Snapshot at 5 ns: initial stage, where the highly compressed quasicylindrical pushing volume $V_p(t)$ expands against the two liquid fronts while the still nearly cylindrical expanding gas volume $V_e(t)$ does it in the radial and axial directions. Approximate illustrating shapes of V_p and V_e are depicted. 15 ns: V_p starts expanding in the radial direction, while V_e begins a doughnut-shaped mixed radial-spherical expansion. 4 μs: V_p undergoes a mixed radial-spherical expansion. 8 μs: both V_p and V_e tend to expand spherically. Final stage ($t \gtrsim 12$ μs): Both V_p and V_e already expand nearly spherically. Supporting pictures from Stan *et al.* [8], Supplemental Material. Positions, shape, and size of control volumes are just illustrative.

blast, or the *efficiency* of the blast against M. The factor $(\gamma - 1)$ can be considered the Grüneisen coefficient of the initial energized matter (in many cases a warm dense matter state [12]), γ being its adiabatic coefficient when it expands. Consistently with the assumption made in Ref. [8], if the evolution of the gas in the blast is assumed quasi-isentropic this coefficient stays nearly constant from large to small densities along the blast, as it will be shown. The first term in the left-hand side of Eq. (1) is the total energy received by the object in the process up to time t. This compact formulation in terms of a single geometrical variable is advantageous for relatively simple geometries like the explosion of a microjet produced by flow focusing [22], which gently carries samples (as initially suggested in Ref. [19], see Supplemental Material [17]): In that application, the microjet is shot by a train of extremely short x-ray pulses in SFX [8,10,23].

Application to the destruction of a liquid cylinder.—Consider a liquid cylinder of diameter d_j where a very large energy density is instantaneously deposited in a slice (see Fig. 2). That energy splits the cylinder (a jet, in SFX) in two symmetrical rods whose separating fronts develop two symmetrical expanding liquid lamellas, whose mechanical energy received from the gas can be expressed as

$$\int_{t_0}^{t}\int_{S_p+S_i} P(\mathbf{x}_p, t')\mathbf{v}_p\cdot\mathbf{n}_p dA dt = \frac{1}{2}\rho_l\frac{\pi d_j^2}{4}\int_{x_0}^{x}\frac{x_t^2}{4}dx = \frac{\pi d_j^2\rho_l}{32}\int_{t_0}^{t}x_t^3 dt, \qquad (2)$$

where x is the distance between the two separating liquid fronts, subscript t indicates the time derivative, and ρ_l is the density of the liquid. x_0 and t_0 are the initial values of the gap size and time, respectively. For conservation of momentum, we assume in expression (2) that the liquid is radially ejected into the two liquid lamellas at an instantaneous speed $x_t/2$ due to the overpressure in V_p.

Besides, the ratio of the liquid thermal layer thickness λ to the jet diameter d_j can be estimated as $\lambda/d_j \sim [K^2 d_j/(\rho_l c^2 E_D)]^{1/2}$, where K and c are the thermal conductivity and specific heat of the liquid. For jet sizes below 100 μm and energies E_D used in SFX experiments, λ would be smaller than the molecular size, which would make the thermal energy transfer to the liquid negligible once the rapid blast takes place. This supports neglecting the internal energy gain by the liquid due to diffusion in Eq. (2). Defining the variables $\phi = x/l_o$, $\tau = t/t_o$, and $\Omega = V_p/(\chi V_o)$ made dimensionless with the characteristic length l_o, time t_o and volume $\chi V_o = l_o^3$ in Eqs. (1) and (2), Eq. (1) can be expressed in nondimensional form as

$$\int_{\tau_o}^{\tau}\phi_\tau^3 d\tau + \chi\Omega(\tau)^{1-\gamma} = \chi, \qquad (3)$$

with its time derivative form as

$$\phi_\tau^3 = \chi(\gamma-1)\Omega^{-\gamma}\Omega_\tau \Rightarrow \phi_\tau^2 = \chi(\gamma-1)\Omega^{-\gamma}\frac{d\Omega}{d\phi}, \qquad (4)$$

where we have defined $t_o = [\pi\rho_l d_j^2 l_o^3/(32E_D)]^{1/2}$. l_o and χ will be determined from dimensional arguments and maximum correlation of experimental data. On the other hand, although $\Omega(\tau)$ is an unknown variable of the problem, the mathematical structure of Eq. (4) and physical principles will univocally fix the asymptotic trends of Ω. Geometrically, ϕ is related to the shape factor of the object (the column) to the effective blast volume, represented by Ω. In the following, their relationships for both large and small times τ are analyzed.

Asymptotic behavior of the pushing gas volume Ω for small and large times τ.—In the very initial stages, both the expanding and pushing volumes V_e and V_p produce an enormous push against the two liquid fronts forming the gap (Fig. 2, time $t = 5$ ns). The geometry of both V_e and V_p remains nearly cylindrical for a while, especially that of V_p. Hence, the nondimensional form of the pushing volume should scale as $\Omega \sim \phi$ since its expansion would proceed predominantly in the axial direction. This occurs because (i) by definition, the expanding volume cannot be radially pushed by Ω at a higher rate than the opening gap, and (ii) the expanding volume pushes against S_s (the periphery of the radially expanding liquid layer, see Fig. 2) with pressures necessarily smaller than those at S_i, preventing a radial expansion of the pushing volume Ω. Therefore, assuming that $\phi \sim \tau^{\alpha_0}$, using Eq. (4) one should have

$$\tau^{2(\alpha_0-1)} \sim \tau^{-\gamma\alpha_0} \Rightarrow \alpha_0 = \frac{2}{2+\gamma}. \qquad (5)$$

On the other hand, in the last stages of the blast (just before surface tension force overcomes the gas pressure, see Fig. 2, $t \gtrsim 12$ μs), one should expect that both the expanding and pushing volumes would expand predominantly in the radial direction, which allows a self-similar solution like the one early analyzed by Wedemeyer [21]. This solution yields the following radial distribution of the total energy inside the gas sphere (including both V_e and V_p):

$$\frac{P}{\gamma-1} + \rho_g\frac{v^2}{2} = P_o\left[\frac{(Bt^{-1})^{3\gamma}}{\gamma-1}(1-\xi^2)^{[\gamma/(\gamma-1)]} + \frac{\gamma}{2B^2}\left(\frac{B}{t}\right)^3(1-\xi^2)^{[1/(\gamma-1)]}\xi^2\right], \qquad (6)$$

where $B = [3^{1/2}(\gamma-1)/2]$, $\xi = [\rho_o B/(\gamma P_o)](r/t)$, ρ_o is the initial density of the expanding gas, v the gas speed, and r the radial spherical coordinate from the center of the gap (Fig. 2. See Supplemental Material [17] for additional details on this solution). The fundamental conclusions from this solution are: (i) For $\xi \ll 1$ (i.e., inside V_p or Ω), the kinetic energy to pressure ratio becomes as small as ξ^2; i.e., pressure dominates over inertia in V_p as anticipated. (ii) According to the self-similar nature of the solution, the position of the expanding edge of V_p would correspond to a constant (small) value of ξ according to mass conservation, for any value $\xi \leq 1$ [21], since the gas should move with a constant speed $v = a_o\xi/B$ at that edge, while $\xi = 1$ is the expanding edge of V_e. Hence, one should expect $\Omega \sim \tau^3$ for $\tau \gg 1$. Again, assuming that $\phi \sim \tau^{\alpha_1}$ and using Eq. (4) one should have

$$\tau^{3(\alpha_1-1)} \sim \tau^{-3\gamma+2} \Rightarrow \alpha_1 = \frac{5}{3} - \gamma. \tag{7}$$

In this limit, one would have $\Omega \sim \phi^{3/(5/3-\gamma)}$. Interestingly, that solution would demand a logarithmic evolution of the gap for perfect monoatomic gases ($\gamma = 5/3$). This is the only scenario for which a logarithmic evolution is contemplated, in contrast to the general logarithmic trend proposed by Stan *et al.* [8].

In summary, one may approximately express the evolution of the gap $x(t) = l_o\phi(\tau)$ as

$$\phi = \phi_o\tau^{\alpha_0}[1 + (\tau/\tau_1)^\delta]^{(\alpha_1-\alpha_0)/\delta}, \tag{8}$$

where constants ϕ_o, τ_1, and δ should be obtained by fitting to either experiments or numerical simulations. Finally, l_o was expected to be proportional to d_j in Ref. [8]. However, having defined $V_p(t_0) = \chi V_o = l_o^3$ and expecting $l_o \sim x_0$, from the definition of the initial energized volume $V_o = x_0(\pi/4)d_j^2$ one can conveniently define

$$l_o = d_j\chi^{1/2}, \tag{9}$$

where the earlier introduced efficiency χ should be a function of the geometry ratio $\eta = r_B/d_j$ of the beam radius r_B to the jet diameter, and the ratio of initial energy density E_D/V_o (see Supplemental Material [17] for SFX) to the energy density of cohesion $\rho_l H_v$ ($\simeq$2.3 GPa for water), that can be written in terms of the parameter:

$$\Pi_v = \frac{\pi r_B^2 d_j F(\eta)\rho_l H_v}{E_D}, \tag{10}$$

where H_v is the heat of vaporization at the temperature of the liquid. $F(\eta)$ is a function of the order unity reflecting the shape of the beam and the influence of the beam to jet ratio (see Supplemental Material [17] for details). Usually, one has $\Pi_v \ll 1$ and therefore a simpler functional dependency as $\chi = \chi(\eta)$ is expected.

To verify our model, we have used the experimental results published in Ref. [8]: fourteen combinations of pulse energies and cylinder (jet) diameters, keeping the liquid (water) and beam focus r_B constant. Figure 3(a) depicts the measurements of the gap distance x as a function of time. We use the properties of water at ambient temperature ($\rho_l = 1000$ kg/m^3, $\sigma = 0.072$ N/m,

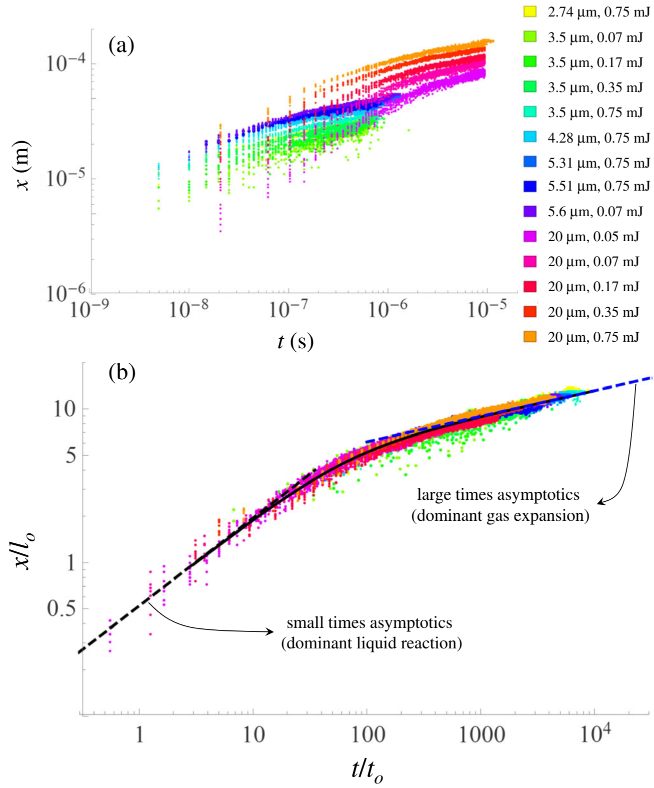

FIG. 3. (a) Dimensional data from Stan *et al.* [8]. Color codes identify the different jet diameters and pulse energies. (b) Optimal data collapse using $t_o = [\pi\rho_l d_j^2 l_o^3/(32E_D)]^{1/2}$ and $l_o = d_j\eta^\beta$ to make times and distances nondimensional. The approximate analytic solution (black line) shows a remarkable fitting to data after optimum collapse. Dot-dashed lines: $\tau = t/t_o \ll 1$ (black, small times asymptotics); $\tau \gg 1$ (blue, large times asymptotics).

$\mu_l = 0.001$ Pa s^{-1}) for Stan's experiments. In their analysis, they readily used d_j as the reference length, and two possible characteristic times, namely, τ_l for a low pressure model, and τ_h for a high pressure model [see Supplemental Material [17], Figs. 2(a) and 2(b)]. In our proposal, defining $\chi = \eta^{2\beta}$ in Eq. (9), one can investigate the role of the beam-to-jet diameter ratio in the characteristic distance l_o.

We have performed a statistical correlation analysis between τ and ϕ data, before the liquid surface tension takes over. We have found that the chi-squared logarithmic differences with local averages is minimized (indicating optimum data collapse: see Supplemental Material [17] for details and quantitative values for the goodness of fit) for $\beta = 0.14 \pm 0.003$ and $E_o = 48.5 \pm 0.5$ μJ, where E_o is the minimum energy at the onset of electrostatic trapping (Supplemental Material [17]). This last value would yield approximately 60 MJ/kg for a beam radius of 0.5 μm and 31 MJ/kg for 0.7 μm, in agreement with the estimation by Stan *et al.* (30 MJ/kg).

Using those previous best correlation values of β and E_o, in what follows we seek the best fit of the approximate function (8) to the experimental data. The minimum chi-squared logarithmic difference is obtained for $\phi_o = 0.523 \pm 0.002$, $\tau_1 = 65 \pm 0.5$, and $\delta = 1.2 \pm 0.05$, with $\gamma = 1.5 \pm 0.01$. This supports the hypothesis in Ref. [8] that the Grüneisen coefficient for water is a nearly constant value $\Gamma = \gamma - 1 \simeq 0.5$ along the process. The approximate function (8) is also plotted in Fig. 3(b) for reference, showing a remarkable fitting. Given the large numerical value of τ_1 in the solution (8), the analysis of large or small values of τ should be obviously understood as compared to τ_1. Moreover, assuming that the evolution starts when the material in V_o is already in dense vapor phase and the initial velocity of the liquid fronts is approximately equal to $H_v^{1/2}$, from Eq. (8) the initial gap would result $\phi_{\text{init.}} = G(\eta)\Pi_v^{-1/\gamma}$, where $G(\eta) = [(\gamma+2)^{-2}\phi_o^{\gamma+2}\pi\eta^{2-\beta}F(\eta)]^{1/\gamma}$. Finally, the efficiency of the explosions resulted as $\chi = (r_B/d_j)^{2\beta} = (r_B/d_j)^{0.28}$, where r_B/d_j range from 0.025 to 0.2 (for d_j from 20 to 2.74 μm). In other cases where $r_B > d_j$ [23], further analysis would provide an extended knowledge of the efficiency χ and the additional verification of the physical insights here proposed. Indeed, using $r_B > d_j$ in SFX would significantly increase the sample hit rate by covering the whole jet section, making measurements insensitive to small accidental drifts of the jet.

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[17] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.123.064501 for (i) a general formulation of mass and energy conservation principles for a blast partially against a deformable object (Appendix A) [18]; (ii) irradiated volume and energy deposited by the x-ray pulse (Appendix B) [19]; (iii) initial effective energy density deposited in the liquid (Appendix C) [20]; (iv) the spherical blast solution of Wedemeyer (Appendix D) [21]; (v) previous nondimensional representations (Appendix E) [8]; and (vi) correlation analysis and optimal fitting (Appendix F).
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