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MASTER
THE THERMODYNAMICS OF THE FUEL FRAGMENTATION GAS *

R. B. Perez
R. G. Alsmiller, Jr.

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In the context of nuclear reactor safety studies, a program is in progress at ORNL¹ whereby fuel-fragmentation situations are mocked up by the application of high-current capacitor discharges through solid UO₂ samples. The goal of the present work is to predict such quantities as the number of gas and liquid fragments and their energy distributions. The point of view adopted in this work is that upon fragmentation, a cloud of UO₂ vapor is formed containing "primeval" liquid fragments which act as condensation centers. In the evolution of time, fragment growth is controlled by nucleation, coagulation and evaporation processes. Eventually, the vapor-droplet system will reach a situation in which clusters (fragments) of various sizes and UO₂ vapor will coexist in an "association-disassociation" equilibrium. Thus, the physical model considered here consists of the identification of the fragmentation gas with an "imperfect" vapor², made up of interacting UO₂ vapor and liquid fragments. The mathematical formulation of the physical model sketched above for the fragmentation gas is based on the quantum mechanical probability distribution function³, properly modified by particle and energy conservation constraints:

$$P(Q|Q) = Z_1^{-1} (2\pi i)^{-1} \sum_{(v)} \left\{ \int d\alpha d\omega \phi_{(v)}^*(Q) \exp[\alpha(E_I - E_{(v)}) + \omega(N_T - \sum_j n_j N_j) - \beta H(Q)] \phi_{(v)}(Q) \right\} \quad (1)$$

where

Q = the set of internal and translational coordinates of the fragments,

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E_I = initial energy in the sample at fragmentation,
 N_T = total number of particles in the sample,
 $\phi(v); E(v)$ = wave function and total quantum energy of the gas in its
 (v) quantum state,
 $\beta = (kT)^{-1}$, (2)
 $k; T$ = Boltzmann constant and temperature of the gas,
 H = the Hamiltonian of the system,
 $Z_1 = Z / \prod_j (N_j !)$, (3)
 $\sum_{(v)}$ = sum over the discrete and continuous eigenvalue spectra,
 Z = the Grand Partition Function,
 N_j = number of fragments containing n_j monomers.

On the basis of equation (1) for the quantum mechanical distribution function we have obtained the following results:

(a) The partition function, Z , equation of state, and the thermodynamic potentials of the fragmentation gas..

(b) The temperature of the gas

$$T = k^{-1} \epsilon_0 \exp[M_k] \quad (4)$$

where ϵ_0 is an energy which depends on the initial energy, E_I , the binding energy per molecule in the sample at fragmentation, u_{LL} , the binding energy per molecule in the liquid fragments, u_L , and the corresponding quantities for the gas fragment, u_G , and where M_k is a "mixing" correction factor arising from the conservation constraints.

(c) To first order, the kinetic energy of the fragments is distributed according to the Maxwell-Boltzmann distribution function.

(d) The size-distribution function is given by

$$P(n_j) = S_n^{-1} n_j^{(4-q)} \exp[-\beta(\sigma n_j^{2/3} - kT \ln R_s)] \quad (5)$$

where S_n , the normalization constant is a function of the temperature and volume of the gas, q is a constant, (the "packing" parameter $q = 3$)², $\sigma n_j^{2/3}$ is the surface energy, and R_s is the saturation ratio, expressed as the ratio of the monomer population partial pressure, p_1 , to the partial pressure of the polymer population.

(e) For supersaturated conditions ($R_s > 1$), there is a critical fragment size, n^* , given by the relation

$$(n^*)^{1/3} = 2\sigma/(3kT \ln R_s) \quad (6)$$

Beyond this critical size, n^* , there is an unstable heterophase transition whereby the "liquid-phase" grows at the expense of the gas fragment population until total "condensation". Calculations were performed for a 10-g sample of UO_2 and a capacitor discharge of 6000 joules. Some of the results are shown in Table I.

Fragment-size distribution functions for the polymer population are shown in Fig. 1. For increasing monomer partial pressures, p_1 , the size-distribution shifts towards larger fragments. For supersaturated conditions the distribution curve increases beyond the critical size as a result of the unstable heterophase transition previously mentioned.

Table I

The Temperature, T, Internal Energy, E_{int} , and Kinetic Energy, E_k ,
 of the Fragmentation Gas as a Function of the Partial Pressure
 of the Monomer Population

p_1 (newton/m ²)	T (°K)	E_{int} (Joules)	E_k (Joules)	E_{kp}^d (Joules)	n ^e (molecules)
5276.1 ^a	4018	-38526	372	.3	2282
5278.2 ^b	4022	-38525	371	.007	1.46310^5
5278.205 ^c	4022	-38525	371	.004	1.905710^5

$u_{LL} = 1.9810^{-18}$ J/molecule; $u_L = 1.9910^{-18}$ J/molecule; $u_G = 1.1510^{-18}$ J/molecule.
 Surface tension = 450 dynes/cm.

^aSubsaturation^bSaturation^cSupersaturation^dKinetic energy carried by the polymers^eMost probable number of molecules in a polymer

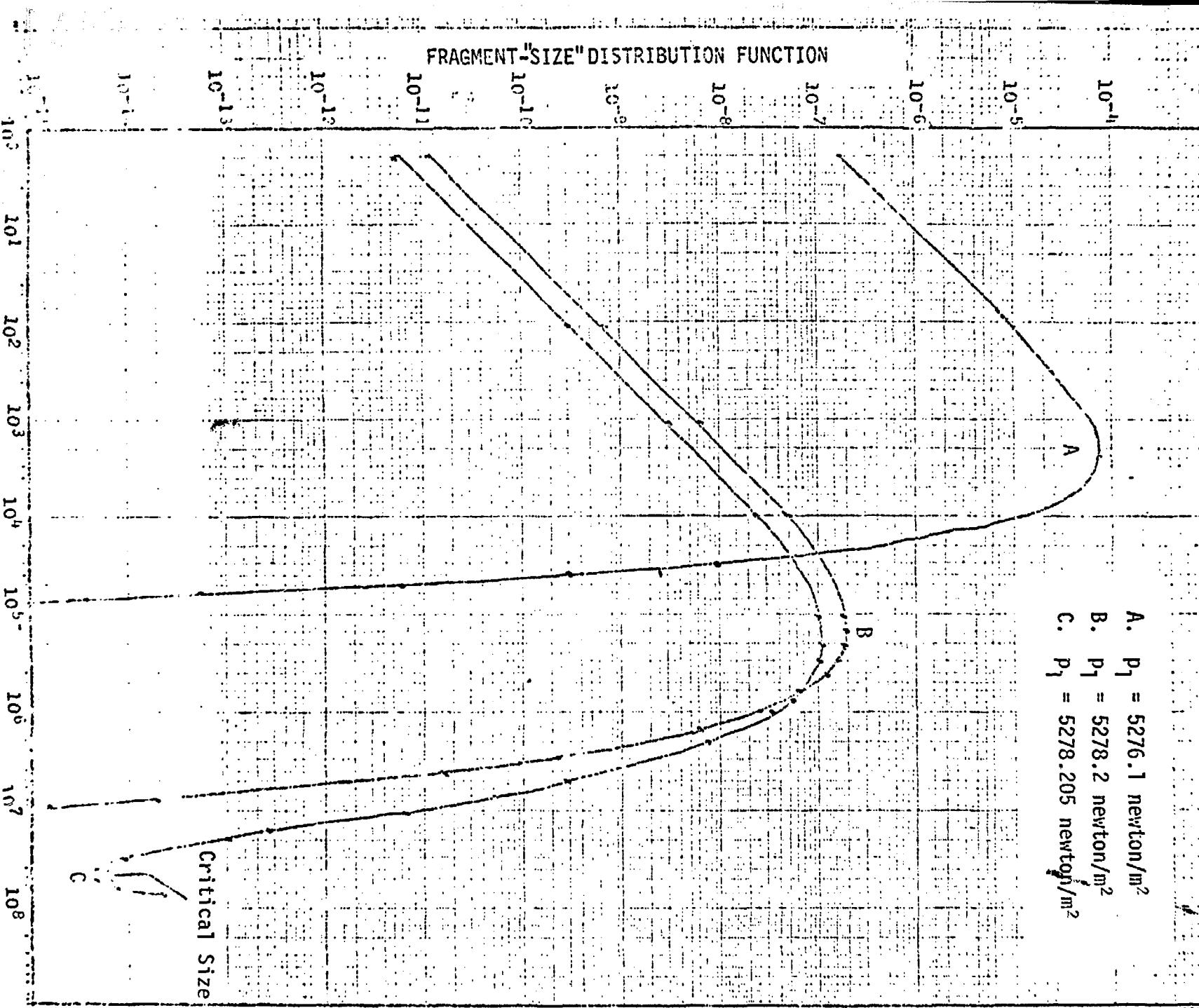


Fig. 1. Fragment-Size" Distribution
as a Function of the Number of Monomers for
Various Values of the Monomer Pressure

References

1. T. S. Kress, G. W. Parker, M. H. Fontana, Oak Ridge National Laboratory ORNL/TM-4875 (1975).
2. F. F. Abraham, Homogeneous Nucleation Theory, Academic Press, New York (1974).
3. J. O. Hirschfelder, C. F. Curtiss, R. B. Bird Molecular Theory of Gases and Liquids, J. Wiley & Sons, New York (1954).