1) Errors from the neglect of polarization may be appreciable when the angles of emission or incidence are such that there is a large difference between the emissivities in the p and s directions.

2) The percent difference between the energy absorbed with and without polarization effects is maximum when there is only one reflection in the system.

3) When externa lincoherent radiation is incident on the system the fraction of energy absorbed by the system is smaller when polarization effects are considered and the maximum difference occurs after some finite number of reflections.

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Fragmentation of Liquid Metallic Particles in Two-Phase Nozzle Flow into a Vacuum

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Introduction

For the generation of Ba ion clouds in space experiments,[1] partial combustion of Ba with CuO in a reaction vessel and subsequent expansion of the liquid reaction products through a nozzle into vacuum is one of the most economical processes. Thermodynamics predicts[2] for an initial temperature of approximately 2500°K, a good yield of Ba vapour, which can be photoionized by the sun's radiation.

When effusing through the nozzle the liquid breaks up into little droplets. The vaporization efficiency from this two-phase jet is then mainly governed by transport phenomena. The droplet size distribution determines: 1) the rate of vaporization, which, in the case of multicomponent droplets (Ba, BaO, Cu), is not so much controlled by the equilibrium vapour pressure, and, e.g., the Hertz-Knudsen equation, as rather by the diffusion of liquid Ba to the droplet's surface[3]; 2) drag, source drag[4,5] and thermal conduction between the polydisperse liquid and the vapour phase, and hence the degree of dynamic and thermal disequilibrium in that part of the jet, where vaporization takes place; 3) recondensation of vapour in the outer parts of the expanding two-phase jet, because the larger the particle size, the sooner recondensation is thermodynamically possible[4] and will take place in jets from larger release payloads; 4) the nonsteady expansion of the vapour from the two-phase jet into the free molecular flow regime; the terminal expansion velocity of the vapour is determined essentially by friction and heat transfer from the particulate phase.[6] Thus, the desire for knowledge and control of the particle size distribution has prompted this laboratory study on the particulate phase of a Ba-vapour jet in a vacuum chamber.

Received December 15, 1969. We would like to thank R. Lüst for supporting this work.

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Experimental Description

1. *Source*

A liquid mixture of Ba, BaO, and Cu at a temperature of 2500°K was obtained by burning Ba and CuO in a molar ratio 2.5:1 in a 50 cm³ reaction vessel. The pressurizing gas, the effect of which on particle atomization was to be investigated, was N_2, which was split off during the reaction from NaN_3 administered to the original reaction mixture in amounts of about 0.4% by weight. Chemical conversion was complete within less than 10 msec[7] and the liquid reaction products were expelled through a convergent tungsten nozzle (diam: 5 mm) into a 20 m³ vacuum chamber, initially at 10^{-5} torr. The mass-flow rate through the nozzle decayed steadily to zero within about 300 msec after ignition.[7] Except for the first precursor,[8] the residence time of the reaction products inside the vessel at a pressure between 20 and 100 atm was long enough to initially establish thermal equilibrium.

2. *Jet boundaries*

The flow lines of particles become source-like early in the expansion with an angular density profile of $\cos^2$-shape,[7] the maximum flow angle, determined with a high-speed camera,[8] being close to $\theta_m = 60°$ and nearly constant throughout the effusion.[7,8] Even though the initial pressure in the chamber was low enough to permit free expansion of the vaporizing two-phase jet, any analysis might have been affected by N_2 reflected from the chamber walls, and by H_2 released from not thoroughly dried surfaces by impinging metal atoms. Prediction of the location of the Mach disk in two-phase jets under these conditions is laden with uncertainties.[9] To exclude such perturbations on the centerline of the jet, where the particle analysis was to be made, the variation of the number density of liquid particles with time was monitored by means of laser-light scattering at a distance of 330 cm from the nozzle.

Because the scattered radiation had to be much more intense than both reflected light from the nozzle (~2500°K) and the brightness of the particles themselves (~1350°K),[8] both the incident light beam from a 80 mw cw He-Ne-laser and the light scattered at an angle of 40° (Fig. 1) were protected by light tubes. To suppress any reflections 7 stops had to be inserted into the tube surrounding the scattered light beam. The recording photomultiplier was equipped with a narrow band interference filter (half-width 20 Å at 6328 Å). At the junction of the tubes a 14 cm² aperture transmitted part of the two-phase jet.

Oscilloscope traces show that it takes about 20 msec from the commencement of the effusion until the first particles pass the laser beam, indicating a front velocity of the jet of about 160 m/sec. A strong increase in particle number density was observed about 250 msec after the effusion had ceased. This is associated with the Mach disk, which degenerates into a pressure wave as the effusion stops and carries fine dust toward the nozzle. Hence, measurements anywhere on the centerline were expected to reflect the free jet properties.

3. *Particle collector*

Particles were collected on a glass plate in the free jet for analysis by micrography and electron micrography. Because solid barium droplets react vigorously with air, it

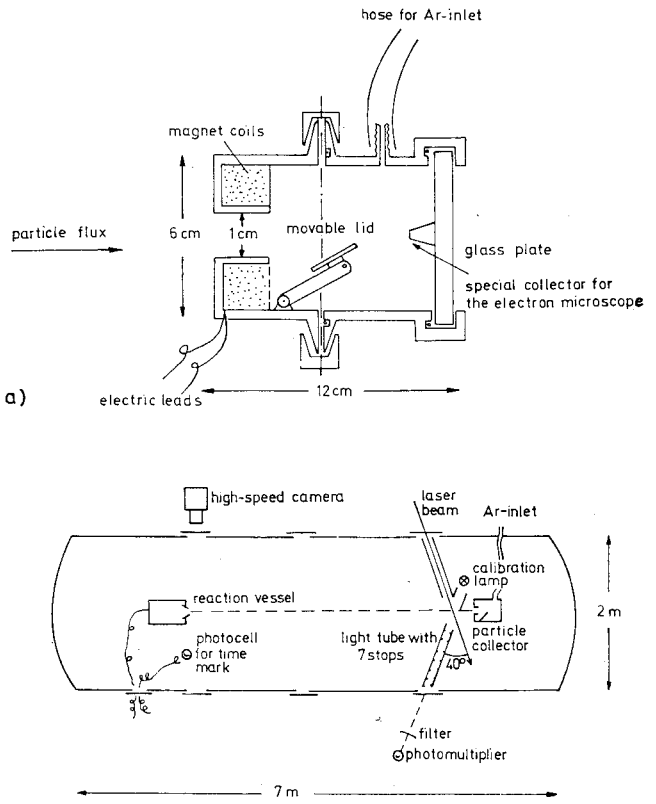


Fig. 1 a) Particle collector. b) Vacuum chamber with experimental setup.

was indispensable to protect the virgin particles from the atmosphere when the chamber was flooded. The particle collector, with solenoid operated shutter and Ar inlet, schematically shown in Fig. 1, was mounted in the vacuum chamber approximately 340 cm from the nozzle. At this distance both gaseous and condensed phases were adequately diluted, so that "skimmer interferences" were negligible, and counting

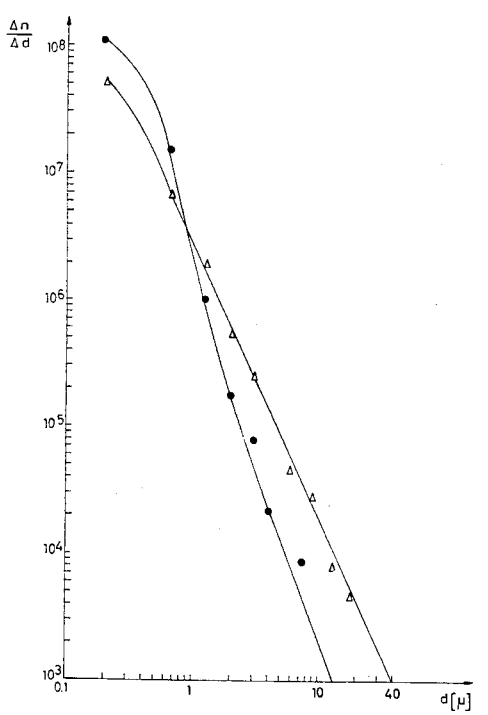
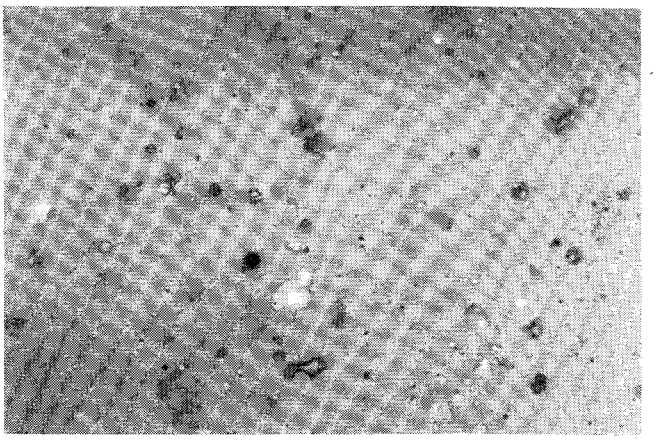
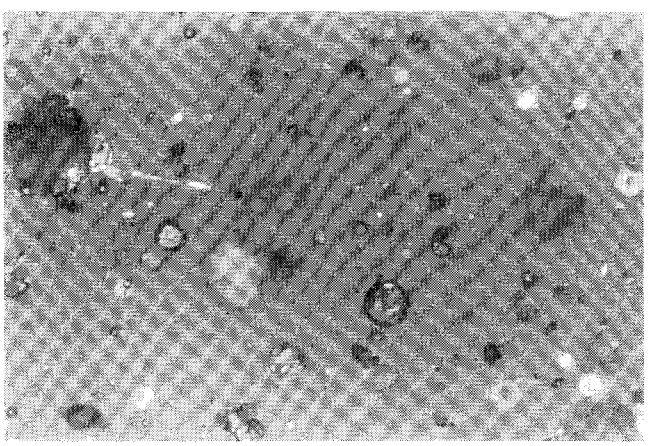


Fig. 2 Particle size distribution; ● addition of 0.4% NaN_3 by weight, △ without addition of NaN_3.



a) Addition of 0.4% NaN_3 by weight



b) Without addition of NaN_3

Fig. 3 Typical micrography of collected particles from two-phase nozzle expansion of burnt Ba-CuO-mixtures (molar ratio 2.5:1). Enlargement × 933.

of individual deposited particles was possible. Because the average particle velocity[7] was above 70 m/sec, the original size distribution was not affected by the deflection of the trajectories by gravity.

At the end of the experiment, the particle collector was closed and filled with high-purity argon; the chamber could then be flooded, and the collector recovered for analysis. A continuous Ar flow was sustained after removal of the upper part of the collector (Fig. 1) during microscopic scan.

4. Microscopic scan

The stereo-microscope (Zeiss) also facilitated determination of the height of the particles. Most of the larger particles were flattened, which indicates that they still were liquid at the time of the impact (melting point of Cu 1357°K, of Ba 983°K). The volume of each particle was calculated by taking diameter and height as major and minor axis, respectively, of an oblate spheroid. The diameter of the equivalent sphere, the shape of the liquid particle before impact, was used for the size distribution.

Submicron particles deposited on carbon foil in the particle collector were analysed by means of an electron microscope (Elmiskop 1 A, Siemens, E = 80 kV, enlargement approximately 160.000).[10] To obtain a representative sample of these particles, sizing and counting of a field of 10^{-5} cm^2 was sufficient, whereas particles in the μ-size range required scanning of 10^{-4} cm^2. Histograms were constructed on the basis of properly chosen size ranges and the number frequencies were normalized to a size range of $\Delta d = 1$ μ and an area of 1 cm^2. The corresponding size frequency curves $\Delta n/\Delta d$ vs d are shown in double logarithmic plots (Fig. 2).

Table 1 Average sizes of collected particles

		without NaN_3	with NaN_3
Arithmetic mean, on number basis	$d_n = (\Sigma fd)$	$= 0.34\ \mu$	$0.27\ \mu$
Surface area, on number basis	$d_s = (\Sigma fd^2)^{1/2}$	$= 0.67\ \mu$	$0.39\ \mu$
Volume, on number basis	$d_v = (\Sigma fd^3)^{1/3}$	$= 1.49\ \mu$	$0.87\ \mu$
Geometric mean, on weight basis	$d_m = \Sigma fd^4/\Sigma fd^3$	$= 13.70\ \mu$	$12.81\ \mu$

The mass deposited per unit area

$$m_D = \frac{\pi\rho}{6}\sum_{\Delta d} d^3\frac{\Delta n}{\Delta d}$$

where the bulk density is $\rho = 4.6$ g cm^{-3}, agreed within 20% with that value calculated from the total ejected mass M and the dilution factor for $\cos^2$ distribution[7] of mass-flow densities over flow angles θ at the nozzle distance x on the centerline;

$$m_D = \left[1\Big/\int_0^{\theta_m}\cos^2\frac{\pi}{2}\frac{\theta}{\theta_m}\sin\theta d\theta\right]\cdot M/2\pi x^2 = 1.02M/x^2$$

This consistency confirms the applicability of the flow picture and that representative samples were obtained also of the larger particles, which are seldom numberwise but still determine the mass weighted average.

With a few exceptions the droplets seemed to be a homogeneous mixture of all three reaction products, as they had uniformly the same reddish colour[8] (determined by Cu) and electron diffractograms did often not reveal any crystal structure,[10] which also is absent in many salt aerosols.[12]

Results and Discussion

By inspection of the two micrographs (Fig. 3) one recognizes that addition of controlled amounts of N_2 produces much finer particles than observed for reaction mixtures, where the N_2 contents depend on accidental impurities (approximately 500 ppm N_2 in commercial Ba and decomposition products from ablative insulation of the reaction vessel). This is quantitatively born out by the two size-frequency curves (Fig. 2), each being constructed with data of many effusion experiments. The various physically relevant average sizes[11] are, with the normalized distribution function

$$f = (\Delta n/\Delta d)/(\Sigma\Delta n/\Delta d)$$

given in Table 1.

These size distributions from this expansion into vacuum indicate a larger abundance of smaller particles than observed in rocket exhaust plumes,[13] in which case small particles are supposed to coalesce to larger ones during the dwell time upstream of the nozzle.[14] By the same token, one may surmise that particle fragmentation of these jets with an initial weight fraction of condensed matter of more than 95% takes place mainly downstream of the nozzle where agglomeration is no more possible.

To be sure, a detailed analysis[7] of the nozzle flow and of the pressure change inside an accelerated reaction vessel has shown, that the liquid is dispersed already before passage through the nozzle, but with much larger mean droplet size than derived from the terminal size distribution reported here.

The fragmentation mechanism is normally sought in shear and turbulence, effects which might only be of minor importance here because of the low Reynolds numbers.[5] The enhancement of atomization upon addition of NaN_3 rather suggests, that N_2 is partially solved by the liquid metal at the high pressure inside the reaction vessel and assists in further droplet break-up upon discharge. The vapor pressure of Ba is probably too low under these conditions to exceed the surface pressure $\Pi \gtrsim 10^6$ dyne/cm^2 of particles with $d \simeq 10\ \mu$. Even though this analysis of droplet sizes has been made without time resolution and only on the centerline of the jet axis (with increasing flow angle a shift of the size distribution toward smaller diameters is expected), it indicates how to control the size distribution by simple means. The large concomitant increase in vapor yield of Ba has been demonstrated elsewhere.[7]

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A Transformation between Axisymmetric and Two-Dimensional Turbulent Boundary Layers

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IN Ref. 1, Mangler gives a method for relating the properties of a laminar boundary layer on a body of revolution to those of a corresponding two-dimensional flow. There did

Received July 22, 1969; revision received January 13, 1970. This investigation supported by Naval Air Systems Command, AIRTASK No. 320/292/69-R009-02-030.

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