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VAPORIZATION OF RADIOISOTOPE FUELS IN LAUNCH VEHICLE ABORT FIRES

David C. Williams
Materials Development and Analysis Division

SANDIA LABORATORIES

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ABSTRACT

Models are developed for estimating the extent to which radioisotopic fuel forms will vaporize if released to the fires resulting from the catastrophic aborts of rocket booster vehicles. Pad aborts of both liquid and solid-propellant systems are considered, and aborts after lift-off are also treated. Estimates are given for the size and activity spectra of the radioactive particles produced when the vapor recondenses. Numerical results are specifically calculated for certain $^{238}\text{PuO}_2$ fuel forms (microspheres, solid-solution cermet, and plutonia-molybdenum cermet), but the models themselves should also be applicable to other refractory actinide oxides. The uncertainties involved in the analyses are discussed in some detail. Out of necessity, many simplifications and mathematical approximations were made in the models employed. However, these quite likely do not result in as much error as do the uncertainties in some of the necessary input data concerning fuel-form properties and abort environment characteristics.

Key words: Aerospace, nuclear safety, SNAP

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Summary

This report reviews all work performed by Sandia Laboratories on the problem of vaporization of radioisotope fuels in fires resulting from rocket booster aborts. The model previously reported (SC-RR-68-441), which describes the vaporization of $^{238}\text{PuO}_2$ microspheres in liquid-propellant pad-abort fires, is generalized to include other radioisotope fuel forms, such as the solid-solution cermet (SSC) and plutonia-molybdenum cermet (PMC). Models are also developed for vaporization after vehicle lift-off (altitude aborts) and for aborts of solid-propellant vehicles. The recondensation of the vaporized material is also analyzed, primarily with respect to the range of particle sizes and levels of radioactivity expected.

If $^{238}\text{PuO}_2$ fuel is in the form of relatively small particles (a few hundred microns in diameter or less), substantial vaporization will result if it is exposed to the full effects of a liquid-propellant pad-abort fireball, even if the vehicle involved is a relatively small one. Cermet-type fuels will vaporize considerably less if they do not suffer excessive degradation to small particles; however, available thermodynamic data suggest that the fireball thermochemical environment could cause rapid disintegration of the metallic (molybdenum) matrix, which would substantially increase the vaporization. In most cases, altitude aborts will result in somewhat less vaporization than launch pad aborts. The situation with respect to solid-propellant fires is found to be rather complex, but in some cases the hazard may be less than for liquid propellants because of the tendency of small particles to be quickly blown out of the fire. After the radioisotope recondenses, most of the activity will be associated with small particles (a few microns in diameter or less) which lie in the respirable size range and which are capable of undergoing atmospheric transport over large distances.

The numerical results given in this review were necessarily calculated for specific combinations of launch vehicles and radioisotopic fuel forms. Some simple scaling laws are offered for extrapolating the results to other situations, but these should be employed only to obtain quick, rough estimates. Any comprehensive analysis should apply the basic models directly to the situation of interest. Hence, the emphasis is upon these models, their range of probable validity, and their application to combinations of vehicles and isotopic fuels other than those specifically considered.

The many uncertainties and possible errors in the analyses must be stressed. The models employed here involved numerous mathematical approximations and assumptions which can result in significant error in some situations. Even more important are the uncertainties as to the correct values of the various parameters required as input to the analysis, both with respect to the characteristics of some of the abort environments and also with respect to the response of fuel-form materials to those environments. Experimental investigation is recommended in certain cases.

I. INTRODUCTION

All major radioisotope thermoelectric generator (RTG) systems currently used (or in advanced stages of development) for space systems employ ^{238}Pu as the basic heat source, either as PuO_2 microspheres or as a PuO_2 -containing cermet in which molybdenum constitutes the metallic phase. In addition, most systems under consideration for the foreseeable future will utilize either ^{238}Pu or some other alpha-emitting actinide in a refractory chemical form. For all these materials, the greatest radiological hazard results when the isotope is dispersed in the atmosphere as a fine particulate in the respirable size range ($\lesssim 10$ microns equivalent aerodynamic diameter^{*}, or $\lesssim 3$ microns true diameter for particles with densities of about ten g/cm^3). Consequently, any process which could reduce the radioisotopic fuel to respirable particulate must be carefully considered. One of the most important of these processes is vaporization of the fuel in the fire resulting from the abort of a launch vehicle.

In a previous report⁽¹⁾ this problem was treated for the specific case of $^{238}\text{PuO}_2$ microspheres exposed to the fireball resulting from the pad abort of a liquid-fueled vehicle with special emphasis on the Saturn V. The purpose of the present work is to extend the treatment to include other types of launch vehicles, other fuel forms, aborts after lift-off (altitude aborts), and the particle size and activity spectrum to be expected after the vaporized material recondenses.

It is manifestly impossible to analyze all the possible combinations of launch vehicles, fuel forms, and abort situations which might be conceived. Consequently, emphasis will be on $^{238}\text{PuO}_2$ microspheres and solid

^{*}The aerodynamic diameter is defined as $d\rho^{\frac{1}{2}}$, where d is the true diameter and ρ is the density.

solution cermet (SSC) or plutonia molybdenum cermet (PMC) fuel forms (those most likely to be used in the near future), and upon the fires resulting from the aborts of Saturn V and Titan III launch vehicles (the largest liquid-propellant and solid-propellant systems, respectively, currently planned for use). Simple approximate scaling laws are offered for other systems, which permit one to obtain rough estimates with a minimum of recomputation. Detailed safety analyses must, of course, always examine the specific system of interest directly.

Section II describes the basic vaporization model used, with those details given in Ref. 1 largely omitted. Sections III, IV, and V deal specifically with liquid-propellant pad aborts, solid-propellant pad aborts, and altitude aborts, respectively. Section VI treats the recondensation problem. In Section VII, some of the uncertainties in this analysis are discussed; none of the results offered here should be used without careful consideration of these uncertainties, as some are substantial.

One final point must be stressed: in no sense is this review to be regarded as a full safety analysis report for the vaporization source term. Questions as to how much fuel might be released to a given abort situation, etc., are completely beyond the scope of this work. Terms such as "fraction vaporized" refer only to the fraction which will be vaporized for whatever amount of fuel is exposed to the environment under consideration. If one curie is released under severe conditions for which the "fraction vaporized" is predicted to be 50%, the total vaporized is still only 500 millicuries; if 100,000 curies are released to a less severe condition for which the "fraction vaporized" is only 1%, the total amount vaporized will still be 1,000 curies. To reiterate: "fraction vaporized" means only the fraction of whatever fuel is released, and not the fraction of the total fuel in the RTG in question.

II. Vaporization Model

The basic vaporization model employed is essentially that given in Reference 1, to which the reader is referred for details. The vaporizing particles were assumed to be spheres with the vaporization controlled by the rate of transfer of vapor away from the saturated boundary layer at the surface. The latter assumption is essentially valid for the cases of interest here, except for small particles in altitude aborts, as discussed in Section V and Appendix A. The assumption of spherical particles is not valid in some cases, but should not lead to any great error if one uses an "equivalent sphere", defined as having the same surface/volume ratio as the actual particle of interest.

With these assumptions, rate of weight loss per unit area, w/A , is given by the semi-empirical relation⁽²⁾

$$\frac{w}{A} = 2 \frac{D}{d} [1 + 0.276 \text{Re}_d^{1/2} \text{Sc}^{1/3}] C_s \quad (2.1)$$

Here D is the diffusion coefficient for plutonium vapors diffusing into the combustion gases, d is the diameter, and C_s is the concentration of the saturated vapor.* The Reynolds number, Re_d , is equal to vd/ν , where v is the velocity of the particle with respect to the surrounding gases and ν is the kinematic viscosity of the combustion gases (defined as η/ρ_g , where η is the dynamic or "ordinary" viscosity and ρ_g is the gas density). Sc is the Schmidt number, equal to ν/D . In most cases, v was taken to be the particle terminal fall velocity. The relation (2.1) neglects the consequences

* A listing of all symbols used in this report may be found in Appendix F.

of turbulence in the combustion gases (see Appendix A for a very brief consideration of this problem).

To obtain the fraction vaporized per unit time, df/dt , we may multiply eq. (1) by the surface/mass ratio, $6/\rho d$, where ρ is the density of the PuO_2 in the fuel form. Doing this, and utilizing the perfect gas law $C_s = p_s M/RT$, we obtain

$$\frac{df}{dt} = 12 \frac{D}{\rho d^2} \left[1 + 0.276 \text{Re}_d^{1/2} \text{Sc}^{1/3} \right] \frac{p_s M}{RT} \quad (2.2)$$

where p_s is the sum of the saturated vapor pressures of all plutonium species present, M is the molecular weight, R is the gas constant, and T is the absolute temperature.

The combustion products for the various systems of interest are complex mixtures of substances, and their composition varies from system to system and also depends upon the temperature. At the temperatures of interest, there is no direct information upon such quantities as ν and D for any of these mixtures. Consequently, estimates were derived by extrapolation from lower temperatures and other materials, as described in Ref. 1. Some error is involved, but probably no more than a factor of two at most.

Except for the vapor pressure p_s , the temperature-dependent terms in equation (2.2) occur in combinations whose net temperature dependence is approximately of the form $T^{0.5 + x}$, $0 < x < 0.5$. Because of the very rapid decline in p_s with decreasing T , only a limited range of temperatures is of interest. Consequently, the other temperature-dependent terms in equation (2) were set equal to their values at some average temperature, usually 2700°K; the resulting error may be shown to be generally under 10%.

When these approximations are made, and numerical values inserted (cgs units) one obtains⁽¹⁾

$$\frac{df}{dt} = 0.079 \frac{p_s}{\rho d^2} \left[1 + 0.1168 (vd)^{1/2} \right], \quad (2.3a)$$

It is convenient to define a quantity Q equal to the time-integral of equation (2.3a):

$$Q \equiv \frac{0.079}{\rho d^2} \left[1 + 0.1168 (vd)^{1/2} \right] \int_{t_r}^{\infty} p_s(t) dt, \quad Q < 1; \quad (2.3b)$$

where t_r is the time the fuel particle is released to the abort fire environment and where the time-variation of all quantities except p_s has again been ignored. Then $f = Q$ for $Q < 1$, and $f = 1$ if $Q \geq 1$.

The vapor pressure as a function of time is, of course, determined by the particle temperature as a function of time, and this, in turn, is governed by the time-temperature history of the abort fire. Since different fire models are required for the different aborts of interest, consideration of these models will be deferred to the relevant sections.

In Ref. 1, it was possible to assume that the $^{238}\text{PuO}_2$ microspheres were always at the same temperature as the surrounding fire, with zero thermal lag. This approximation would be very poor for some of the cases of interest here. Consequently, the temperature response of the vaporizing particles was calculated by numerical integration of the equation

$$\begin{aligned} \frac{dT}{dt} &= \frac{\text{net incident radiant heating}}{\text{particle heat capacity}} \\ &= \frac{\pi d^2 \epsilon \sigma (T_f^4 - T^4)}{\frac{1}{6} \pi d^3 \rho c_p} = \frac{6 \epsilon \sigma}{c_p \rho d} (T_f^4 - T^4), \end{aligned} \quad (2.4)$$

where T_f and T are, respectively, the fireball and particle temperature as functions of time, ϵ is the particle emissivity, σ is the Stefan-Boltzmann constant and c_p is the specific heat. Equation (2.4) is based upon the infinite thermal conductivity approximation; this is adequate except for large pieces of fuel, which undergo little vaporization in any case. A more important limitation is that Equation (2.4) assumes heating by radiation only and neglects heating by conduction from the combustion gases. The latter can be important for the smaller particles (say, $d \lesssim 0.02$ cm; see Appendix B). For these particles, it was usually assumed that heating would be instantaneous as in Ref. 1.

For pure $^{238}\text{PuO}_2$ fuel forms, (e.g., microspheres), the vapor pressure was assumed to be that given by Ackermann *et al.*⁽³⁾:

$$\log_{10} p_s = (7.50 \pm 0.15) - (29260 \pm 280)/T \quad (2.5)$$

The emissivity was estimated to be about 0.5, primarily from comparison with values quoted⁽⁴⁾ for similar oxides such as ZrO_2 and ThO_2 . Other fuel properties were assumed to be $\rho = 10 \text{ g/cm}^3$ and $c_p = 0.09 \text{ cal deg}^{-1} \text{ g}^{-1}$, as estimated from the Mound ^{238}Pu fuel-form data sheets⁽⁵⁾. For the composite fuel forms, SSC and PMC, the vapor pressure was assumed to be reduced by a factor equal to the dilution factor of the $^{238}\text{PuO}_2$ in the composite. Other properties of composite fuel forms, as estimated from data compiled for the SSC at Los Alamos Scientific Laboratory⁽⁶⁾, were assumed to be: $\epsilon = 0.65$, $\rho = 10.5 \text{ g/cm}^3$, and $c_p = 0.083 \text{ cal deg}^{-1} \text{ g}^{-1}$. All these fuel properties are actually weakly temperature-dependent, which was not taken into account in the calculations; the values listed here are in effect "averages".

In brief, then, the fractional vaporization was calculated by estimating $T_f(t)$ from an appropriate abort fire model, determining T as a function of time from equation (2.4), employing equation (2.5) to obtain $p_s(t)$, and inserting the result in equation (2.3b).

III. Liquid Propellant Pad Aborts

Abort fires are violent "unprogrammed" events, and considerable variability must be expected. There is no way to precisely model the resulting fireball. The most common approach is to make those assumptions which maximize the resulting thermal environment. These include release of all propellants prior to the time when the fireball lifts off the ground, complete mixing of all released propellants with their participation in the fireball, complete thermochemical equilibrium, and cooling by radiation only, with no mixing with the surrounding air. The resulting models are expected to give an upper limit to the severity of the actual environments to be expected, but it is also believed that actual aborts could approach this limit. The available data tend to support these expectations.

The fireball model employed here is the one developed by Van Nice and Carpenter⁽⁷⁾ for the abort of Saturn I and Saturn V launch vehicles. The time-temperature history predicted for a Saturn V abort is shown in Fig. 1. More recently, Bader et al⁽⁸⁾ have developed a fireball model incorporating a theoretically defined lift-off time, which the older model did not. The newer model gives a very slightly less severe environment, but the differences between the two are much less than the many uncertainties involved, and the two models should be regarded as being essentially in agreement. The present work employed the older model primarily because some of the calculations were done before the other became available, and the minor differences did not justify recalculation.

Reference 1 reported in some detail calculations for $^{238}\text{PuO}_2$ microspheres with the size distribution of the SNAP-27 fuel (log-normal, $\ln \sigma_g = 0.315$, mass median diameter (MMD) = 127 microns), released in a Saturn V abort fireball. For convenience, the results are reproduced in Fig. 2, with the

Figure 1. Saturn V fireball temperature as a function of time after abort initiation (from Ref. 7).

Figure 2. Fractional vaporization of $^{238}\text{PuO}_2$ microspheres released to a Saturn V fireball, as a function of the time interval, t_r , between abort initiation and microsphere release (from Ref. 1).

fraction vaporized, $\bar{f}$, plotted against the time of release t_r , where $t = 0$ is taken to be the time of fireball initiation.

In calculating the vaporization of SSC or PMC, the crucial factor is the degree of size degradation of the initial fuel form. The undamaged fuel was assumed to be in the form of discs, 0.538 cm thick by 5.46 cm in diameter. In order to analyze the vaporization of an intact disc, the temperature response over the surface was calculated using the two-dimensional heat transfer code CINDA⁽⁹⁾. Effects of finite thermal conductivity and the variation with temperature of the material parameters were included in this one case. Vaporization was then calculated by substituting an "equivalent sphere" having the same surface/volume ratio as the disk, and a temperature equal to the hottest part of the disk, i.e., the corners.

In any actual accident situation, fuel will not be released unless the capsule containing it is breached by melting or violent insult such as impact or fragment penetration. In the latter cases, at least, the fuel will very likely be fragmented and pulverized, especially near the point where the capsule is breached. Consequently, a wide range of particle sizes must be considered. Calculations were therefore run for SSC (or PMC) vaporization in a Saturn V fireball as a function of particle diameter. Equations (2.3) - (2.5) were employed which means that the effects of finite thermal conductivity and the temperature-dependence of the material properties were ignored. It was also assumed that the released particles were not additionally degraded in size by the attack of the fireball thermochemical environment upon the molybdenum matrix; failure of this assumption could have serious consequences, as will be seen below.

Results are given in Figs. 3 and 4. In Fig. 3, the fraction vaporized is plotted against particle diameter, and the numbers in parentheses are the

Figure 3. Vaporization of $^{238}\text{PuO}_2$ -containing ceramic or cermet fuels in a Saturn V fireball as a function of particle size for the values of t_r given in parentheses.

Figure 4. Vaporization of $^{238}\text{PuO}_2$ -containing ceramic or cermet fuels in a Saturn V fireball as a function of t_r for the particle sizes indicated by the figures on the right-hand side.

time of release, t_r . In Fig. 4, the roles of $\underline{d}$ and t_r are reversed.

Clearly, the vaporization declines very rapidly with increasing particle size, and the amount of vaporization will be largely governed by the amount of SSC or PMC that is reduced in size to a few hundred microns or smaller.

In Fig. 5, the fraction vaporized is given for the particle size spectra that resulted from two different impact tests (Table 3.1)⁽¹⁰⁾. These spectra cannot be directly applied to actual aborts, but qualitatively they are characteristic of what must be expected in that they show a very wide range of sizes, from large pieces down to $d < 10^{-3}$ cm. Also shown is the fractional vaporization calculated for the size spectrum resulting from oxidation of the molybdenum matrix, which might occur quite rapidly within the fireball. If so, it is clear from Fig. 5 that the vaporization could be greatly enhanced.

For an intact, undegraded fuel puck, the fraction vaporized is predicted to be only $\sim 0.01\%$.

The model employed here could be applied to the abort of any other liquid-propellant vehicle, assuming an abort fireball model were available. Rather than considering additional vehicles individually, however, we shall instead obtain a generalization of the preceding results. To do so, we must generalize the fireball model, which necessarily introduces an additional degree of approximation.

As noted earlier, it was assumed that the fireball cools by radiation only. Hence, the cooling rate is proportional to the surface/volume ratio, which varies approximately as $W^{-1/3}$, where W is the weight of propellants involved. Roughly speaking, then, all fireballs should evolve through a similar time-temperature history, with the time scaling as $W^{1/3}$. That is,

TABLE 3.1

Particle size distribution resulting from fuel disk degradation.

<u>Diameter Range</u>	<u>Fraction in Size Range</u>		
	<u>Impact Tests</u>		
<u>(microns)</u>	BMI AA-61 <u>(278 fps)</u>	BMI AA-64 <u>(325 fps)</u>	After Molybdenum <u>Oxidation</u>
> 2000	0.9282	0.5821	-
841-2000	0.0519	0.2103	-
420- 841	0.0051	0.0789	-
177- 422	0.0020	0.0442	0.0870
125- 177	0.0010	0.0081	0.2183
74- 125	0.0006	0.0163	
44 - 74	0.0006	0.0130	0.2456
30 - 44	0.0010	0.0183	0.3660
20 - 30	0.0012		
10 - 20	0.0025	0.0141	0.0728
< 10	0.0015	0.0147	0.0102

Figure 5. Vaporization of $^{238}\text{PuO}_2$ -containing ceramic or cermet fuels in a Saturn V abort for the particle size spectra listed in Table 3.1.

fireballs of different sizes should be at approximately equal temperatures at equal values of the "reduced time" $t/W^{1/3}$, and it is this assumption which we will make here.

Before continuing, however, it must be stressed that this is far from a precise law of nature. Other significant factors such as propellant release rate need not scale as $W^{1/3}$. According to the model of Bader et al (8), the time of fireball lift-off (which affects subsequent fireball evolution) should scale as $W^{1/6}$, not $W^{1/3}$. Furthermore, different propellant mixtures have different energy contents. Since vaporization is so strongly dependent upon the temperature, it is especially important to develop an individual fireball model for any propellant combinations having an adiabatic flame temperature which differs significantly from the value (3000°K) calculated for the Saturn V propellant combination^{7,8}.

Using the $W^{1/3}$ scaling assumption, a generalized vaporization model may be developed in three stages as follows:

(1) First, we define a "zero lag vaporization integral" I_0 , equal to the value which the integral $\int_{t_r}^{\infty} p_s(t)dt$ in equation (2.3b) would have if the particle temperature followed the fireball temperature exactly. Because of the $W^{1/3}$ scaling, I_0 is proportional to $W^{1/3}$ for equal values of $t_r/W^{1/3}$. Hence, a plot of $I_0/W^{1/3}$ vs. $t_r/W^{1/3}$ yields a single curve valid (within our approximation) for all fireballs. This curve is given in Fig. 6, with W expressed in pounds.

(2) Next we define a "thermal lag reduction factor" R_e equal to the factor by which the actual value of $\int_{t_r}^{\infty} p_s(t)dt$ is reduced below I_0 because the actual particle temperature lags behind the fireball temperature. From Equation (2.4), the heating rate varies as d^{-1} , while the time available for heating during corresponding periods of fireball evolution varies as $W^{1/3}$.

Figure 6. Values of $I_0/W^{1/3}$ as a function of $t_r/W^{1/3}$, for use with the generalized liquid-propellant vaporization model, where $\underline{W}$ is the weight of propellant involved in pounds.

Assuming a given initial temperature, then, the reduction factor will be the same for equal values of $d/W^{1/3}$ and $t_r/W^{1/3}$. Fig. 7 gives the reduction factor as a function of $t_r/W^{1/3}$ for the values of $d/W^{1/3}$ indicated in parentheses, calculated assuming that the initial temperature of the radioisotopic fuel is 650°K. For radically different initial temperatures, the reduction factors would have to be recalculated. Because of the neglect of heat transfer by conduction from the gases, the thermal lag is probably overestimated for $d \leq 0.02$ cm; the reduction factor should be set equal to unity for these particles.

(3) There will be an enhancement factor E due to convection, i.e., the relative motion of the particle and the fireball gases (bracketed term in Equation (2.3)). This factor is almost independent of fireball properties and is plotted as a function of particle diameter in Fig. 8, where the relative velocity $\underline{v}$ has been assumed to be equal to the terminal fall velocity.

For a given particle size and a given propellant inventory, these three quantities can be read from Figs. 6-8, and the fraction vaporized is then given by

$$f = Q = 0.079 I_o \cdot R_l \cdot E / (pd^2), \quad Q < 1; \quad (3.1)$$

and $f = 1, \quad Q \geq 1.$

The curves in Figs. 6-8 were calculated using the material properties quoted for the solid-solution cermet fuel form. Other $^{238}\text{PuO}_2$ fuel forms should not differ greatly provided any additional constituents present do not interact with the PuO_2 in a way that alters its vapor pressure, other than by simple dilution. For fuel forms based upon other actinide oxides such as Cm_2O_3 , a very similar approach could be used but the various curves would have to be recalculated, especially $I_o/W^{1/3}$.

Figure 7. Thermal lag reduction factors for use with the generalized model as a function of $t_r/W^{1/3}$, plotted for the values of $d/W^{1/3}$ given in parentheses.

Figure 8. Enhancement factor due to relative motion between the particles and the fireball gases, for use with the generalized model.

The use of the generalized model is best explained with the aid of a simple example. Let us assume that a vehicle with a propellant inventory $W = 2 \times 10^5$ lb undergoes a launch-pad abort and that capsule failure by shrapnel penetration can result in immediate fuel release ($t_r = 0$), but that capsule failure by melt-down could not lead to fuel release before $t_r = 4$ seconds. We consider particles with $d = 0.02$ cm.

For the first case, $t_r/W^{1/3} = 0.0$ and, hence, from Fig. 6, $I_0 = 5.5 \times 10^{-5} W^{1/3}$. The value of $d/W^{1/3}$ is 3.4×10^{-4} , and interpolating approximately from Fig. 7 gives $R_l = 0.84$; Fig. 8 shows that $E = 1.25$ for particles of this size. When combined in equation (3.1) these factors give f

$$f = Q = 0.079 \times (5.5 \times 10^{-5} \times (2 \times 10^5)^{1/3}) \times 0.84 \times 1.25 / (10 \times (0.02)^2) \\ = 0.067.$$

With $t_r = 4$ seconds, $t_r/W^{1/3} = 0.0684$, and from Fig. 6, $I_0 = 1.9 \times 10^{-7} W^{1/3}$. Interpolating from Fig. 7 again gives $R_l = 0.62$, and as before $E = 1.25$. With these factors, equation (3.1) then gives $f = 1.7 \times 10^{-4}$.

Some additional estimates of fractional vaporization are tabulated in Table (3.2). They were obtained from the generalized model in the same manner as was illustrated above. These results illustrate the important point that vaporization resulting from prompt release ($t_r = 0$) declines only slowly with decreasing W (roughly as $W^{1/3}$). However, the decline with increasing t_r is much more rapid for small vehicles than for large ones, and to a considerably lesser extent, the same is true for the decline with increasing particle size. It should also be noted that the factors R_l and E , which were almost negligible in the example cited above, become quite important for the larger particle sizes.

TABLE 3.2

Approximate fractional vaporization for various parameter values,
as estimated from the generalized model

W(lb)	d(cm)	t _r (sec)	f
5.5×10^6	0.01*	0	0.80
		4	0.15
	0.2	0	0.0044
		4	0.00085
2.0×10^5	0.01*	0	0.26
		4	0.0008
	0.2	0	0.00053
		4	Neg.

*_R set equal to 1.0

IV. Solid Propellant Pad Aborts

The abort of a solid propellant vehicle does not produce a fireball of large volume, as in the case of a liquid-propellant booster. Instead, burning pieces of propellant grain may be scattered about the launch site and continue to burn on the ground. The resulting fires are of much smaller volume than the liquid-propellant fireball and may be at slightly lower temperatures; however, they can last much longer, up to several minutes.

If a radioisotope fuel capsule is breached in the abort, pieces of the isotopic fuel may be released and fall upon the burning propellant. In addition, the fuel capsule itself may land in the fire and subsequently fail due to the thermochemical environment. In either case, vaporization of fuel can occur.

Estimating the vaporization requires some kind of a model for the fire. Donaldson and Hardee⁽¹¹⁾ have developed fire models based upon the assumption that thermochemical equilibrium exists at all times. These models yield very high flame temperatures, in excess of 3000°K for some propellants. However, heat flux measurements in actual propellant burn tests at this Laboratory⁽¹¹⁾ have never yielded results which approached the very high values implied by these temperatures. One probable reason is the failure to achieve chemical equilibrium. For example, the propellants tested were aluminum-loaded and substantial quantities of unburned aluminum were generally found after the tests; there would be no unburned aluminum if chemical equilibrium had been obtained.

In the present work, calculations were based upon the Titan III propellant UTP-3001. Experimental heat flux measurements for this propellant were as high as any that have been studied. Donaldson has noted⁽¹¹⁾ that heat flux measurements taken at appreciable distances above the surface

during the UTP-3001 test yield peak values of about $200 \text{ BTU/ft}^2 \text{ sec}$ when extrapolated back to the surface; the actual experimental results obtained near the surface were not that high. He recommends that the extrapolated value be used for analysis.

Two fire models have been employed here. In the first, dubbed the "semi-empirical" model, the recommendations of Donaldson were followed and a maximum heat flux of $200 \text{ BTU ft}^{-2} \text{ sec}^{-1}$ was assumed, corresponding to a peak temperature of 2515°K a short distance above the burning surface. In the other model, the adiabatic flame temperature (about 3080°K)⁽¹¹⁾ was assumed. At greater heights above the surface, the temperature was assumed to be determined by radiative cooling of the rising combustion gases.

The resulting temperature profiles as adapted from Ref. 11, are shown in Fig. 9. The temperature is a function of the dimensionless parameter H/L , where H is the height above the surface and L is a "characteristic dimension" of the burning surface (its diameter, if the surface is circular).

With the temperature known, the vaporization rate can readily be calculated as a function of particle diameter from equations (2.3) and (2.5) if the gas velocity can be determined. The latter was estimated by calculating the volume of gases generated per unit area per second and setting the results equal to the velocity; see Appendix C. The propellant burn rate was taken to be 0.06 inches/sec , as observed in tests at this Laboratory⁽¹¹⁾. These assumptions yield calculated gas velocities of about $2 \times 10^3 \text{ cm/sec}$. The calculated vaporization rate (fraction vaporized per second) is plotted against d in Fig. 10 for the semi-empirical and the adiabatic models. Thermal lag has been ignored since the time required to heat the fuel is short compared to the fire duration and, in any case, the fuel temperature already may be close to the fire temperature if release is due to capsule thermal

Figure 9. Solid Propellant (UTP-3001) flame temperature profiles as a function of dimensionless height above the burning surface, H/L , for the adiabatic and semi-empirical fire models.

Figure 10. Vaporization rate, expressed as the fraction vaporized per second, of $^{238}\text{PuO}_2$ fuels in a solid-propellant fire as a function of particle size.

failure. The adiabatic model is seen to give vaporization rates over 100 times greater than the rates predicted by the semi-empirical model.

Vaporization was also calculated for an intact SSC or PMC disk as a function of time in the fire, using the approximations discussed in Sections II and III. The disk was assumed to be at a temperature of 1000°K when released to the fire. Results are plotted in Fig. 11. The vaporization becomes large only if the adiabatic model is assumed.

So far, the duration of exposure to the fire has been treated as an arbitrary parameter for particles of all sizes. Actually, however, it must be remembered that a solid propellant fire is quite turbulent. As previously mentioned, mean gas velocities of the order of 2×10^3 cm/sec are expected, and turbulence may result in higher transient velocities. Consequently, it is highly probable that small particles will be quickly blown out of the fire. Indeed, it is difficult to see how a small, isolated particle can hang suspended in the hottest part of the fire as has been assumed in the preceding calculations. Even if the particle somehow manages to cling to the surface of the burning propellant, it will not experience the full thermal environment because there must be a cooler boundary layer at the solid surface, which cannot be very hot.

In order to obtain a more realistic picture of the vaporization of radioisotopic fuels in a solid propellant fire, a model was developed in which it was assumed that the smaller particles were blown out of the fire as suggested above. This "blow-off" model is described in more detail in Appendix C. In brief, it was assumed that if a particle was small enough so that its terminal fall velocity in the combustion gases was less than the gas velocity, the drag on the particle would exceed the particle's weight and that the particle would be carried away by the gas stream. By

Figure 11. Vaporization of intact $^{238}\text{PuO}_2$ -cermet disks in a solid-propellant fire as a function of the duration of the exposure to the fire. Fuel disks were assumed to be 0.538cm thick by 5.46cm in diameter.

calculating the drag force upon the particle, the rate of acceleration away from the surface could be estimated. After making certain approximations, the resulting equation of motion was integrated to obtain the particle distance above the surface as a function of time. Using the flame profile curves given in Fig. 9, this was converted into a time-temperature history for the particle's environment. The value of $\underline{L}$ (the size of the piece of burning propellant) was set equal to one meter.

The model predicts that particles with $d \lesssim 0.09$ cm are removed from the fire, and that the smaller they are, the more rapidly they are removed. Small particles ($d \lesssim .01$ cm) blow off so rapidly that, even for these, thermal lag effects may not be negligible, unlike the case for liquid-propellant fireballs. However, the thermal response of the particle cannot be calculated from equation (2.4), because heat conduction from the surrounding gases, and not radiation, is probably the predominant mode of heat transfer for very small particles (Appendix B). This is especially true if the combustion gases contain large amounts of hydrogen, which would increase the thermal conductivity. Thermochemical equilibrium calculations imply the presence of considerable hydrogen; however, as already noted, equilibrium does not appear to exist and the actual hydrogen concentrations may be quite different. In addition, it is quite uncertain as to what the temperature of the particle will be when it is released directly to the fire.

In view of the many uncertainties, it did not appear worth while to attempt a quantitative calculation of the thermal response including both radiative and conduction heating. Instead, it was assumed that the particles were at the fire temperature when released. Using the time-temperature history of the particle environment, obtained as outlined above, the

vaporization was calculated from equation (2.3) much as before. This procedure will over-estimate the vaporization if thermal lag effects are significant.

Results are given in Fig. 12, with the fraction vaporized plotted against particle size for both the semi-empirical and the adiabatic flame temperature model. The break between 0.05 and 0.1 cm represents the dividing line between particles which blow off and those which do not. For the small particles, the duration of the exposure to the fire is determined by the rate they blow off and, hence, this time is not a free parameter; for the larger particles, the duration is a free parameter and results are shown for durations of 10 seconds and 300 seconds.

Figure 13 gives the results for both the semi-empirical fire model and the adiabatic fire model when the size distributions in Table I are assumed. Fraction vaporized is plotted against duration of exposure to the fire. The dependence upon duration is, of course, due only to the larger particles which do not blow off. These are seen to dominate the vaporization for times longer than a few seconds. Also shown are the vaporization calculated for microspheres, assuming both the as-produced size spectrum and the spectrum found after a severe capsule impact test (317 ft/sec on granite). All microspheres, and all the oxidation-residue particles, lie in the blowoff range. The fraction vaporized is seen to be considerably smaller for microspheres than for the SSC (or PMC) if exposure of the latter to the fire is prolonged. It should be remembered, however, that the relative amounts of these two fuel forms which are likely to be released from a capsule and thus exposed to the fire may turn out to be quite different; consideration of that problem is beyond the scope of this paper.

Figure 12. Vaporization of $^{238}\text{PuO}_2$ fuels in a solid-propellant fire as a function of particle size, assuming blow-off of small particles as described in the text. Particles too large to blow off were assumed to be exposed to the fire for either 10 seconds or 300 seconds, as indicated.

Figure 13. Vaporization of $^{238}\text{PuO}_2$ fuels in solid propellant fires, assuming blow-off of small particles, calculated for the size spectra listed in Table 3.1, and also for undamaged microspheres and post-impact microsphere size spectra.

Because of the complexities and uncertainties involved in the subject of vaporization in solid propellant fires, it does not seem possible to present a "generalized" model even to the extent it could be done for the liquid propellant case. However, some very crude scaling factors can be obtained by noting that the vaporization is roughly proportional to the vapor pressure and also to the duration of exposure to the fire. The vapor pressure is determined by the temperature and by the nature of the fuel form. The duration of exposure is determined by the time required for blow-off for small particles, which is very approximately proportional to L/v_g for particles of a given size, where v_g is the combustion gas velocity. For particles which do not blow off, the exposure time is presumably equal to the time between release and the time of propellant burnout.

The preceding analysis of vaporization in solid propellant fires involved a number of uncertainties and approximations in addition to those required for the liquid-propellant treatments. Most of the extra uncertainties of which this writer is aware are such as to make the analysis tend toward overestimating the vaporization. The semi-empirical fire model, however, could be in error in either direction. The adiabatic model can only be an over-estimate. Indeed, the latter has not even been used in a fully self-consistent way here, since its temperatures exceed the melting points of the fuel materials involved and no allowance was made for the consequences of this fact.

The most reasonable estimates are probably those obtained by use of the blow-off model in conjunction with the semi-empirical fire model (solid curves, Fig. 12). Even these results must be taken as an order-of-magnitude thing at best. This entire analysis has been offered only in the belief

that calculations based upon any remotely reasonable physical model will still be superior to the only alternative: going by intuitive feelings.

V. Altitude Aborts

If a catastrophic abort of a rocket booster occurs after liftoff, the situation differs in certain ways from the pad abort case. In particular, there is no collecting surface for the reacting propellants; the propellants may be released with a relatively large forward velocity with respect to the surrounding air; and the ambient pressure may be significantly less than one atmosphere.

Since aborts of solid-propellant boosters do not result in a large-volume fireball, significant exposure of radioisotopic fuel to the resulting fire would appear to depend upon the presence of a collecting surface upon which the burning propellant and the fuel can both rest. In an altitude abort, there is no such collecting surface. We therefore conclude that altitude aborts of solid-propellant vehicles present relatively little vaporization hazard, and will consider only liquid propellant aborts for the remainder of this section.

If radioisotopic fuel is released into the fire, or "firestream", resulting from an altitude abort of a liquid propellant vehicle, there is some uncertainty as to just how long the fuel particles will be exposed to the fire. If the abort is accompanied by explosions violent enough to demolish the tankage and release all propellants within a few seconds, the resulting fire would probably have a duration similar to that resulting from a ground abort were it not for the fact that the propellants are released with whatever forward velocity the vehicle has achieved. If this velocity is large, mixing with the ambient air will probably be enhanced, cooling the fireball more quickly.

It has been suggested^(12,13) that the tankage might sustain only moderate damage, permitting the propellants to spill more gradually and that, with

favorable mixing of propellants, there could result a sort of prolonged torch effect giving high temperatures lasting considerably longer than in a pad abort fireball. However, it does not seem likely that fuel particles can be exposed to the full duration of such a prolonged firestream, for several reasons.

Radioisotope vaporization cannot begin until fuel particles are released from the capsules. Once they are released, small particles (say, $\lesssim 250$ microns) will be constrained to follow the motion of the combustion gases produced by the propellants into which they are initially released. These combustion gases will stream away from the "torch" as new propellants are released (motion pictures of altitude aborts confirm this), carrying the particles with them. Since the amounts of propellants involved at any one time are smaller, these combustion gases should cool more rapidly than in the case of the pad abort fireball, where the propellants are released in a time of at most several seconds.

Larger particles need not follow the motion of the combustion gases so closely, but once they are unconstrained they will still follow trajectories of their own and leave the combustion zone in a few seconds or less. There is simply no plausible mechanism for holding an unconstrained, freely-suspended fuel particle in the high-temperature part of a torch-like firestream. Even if some fuel lodges in a part of the vehicle structure and is therefore constrained to remain in the firestream, the same structure will shield it to some extent and the resulting temperature will not be so high.

Two situations were considered here. In the first, it was assumed that the propellants were released abruptly, while in the second a more gradual release was assumed. The dividing line between the two is, of course, vague. Roughly speaking, an "abrupt" release is one in which the

distance traveled by the vehicle during propellant release is less than the diameter of the pad-abort fireball (somewhat over 1,000 feet for a Saturn V). The details of the treatments are given in Appendix D.

Abrupt propellant release:

No separate, detailed calculations were made for this case. Instead, a crude attempt was made to extrapolate the results for the pad abort fireball to higher altitude, taking into account certain effects due to the reduced ambient pressure. Briefly, values of Q in equation (2.3) that were calculated for the pad abort case were adjusted for (a) the reduced vapor pressure due to the lower temperature, and (b) the increased diffusion coefficients D due to the lower gas density. Thus,

$$\frac{Q(h)}{Q(0)} \approx \frac{p_s(h)}{p_s(0)} \cdot \frac{D(h)}{D(0)} \quad (5.1)$$

Here, the argument h is the altitude, and $p_s(h)$ is the PuO_2 vapor pressure (equation 2.5) at the peak fireball temperature for altitude h . This temperature was taken to be the adiabatic flame temperature for the Saturn V propellant combination as calculated by the ACE computer code⁽¹⁴⁾, at the ambient air pressure $P(h)$. The diffusion coefficient $D(h)$ was assumed to be inversely proportional to $P(h)$. Equation (5.1) ignores certain other altitude-dependent factors, but they are relatively minor and tend to compensate for each other (Appendix D).

When the terms in Equation 5.1 are evaluated, it turns out that the increase in D with h virtually cancels the decrease in p_s , leaving Q almost unchanged. Consequently, it appears that use of the results for the pad abort fireball will give a reasonable estimate for the upper limit of the vaporization in the case of an abort after lift-off in which the propellants

are released abruptly. Since the effect of the vehicle's forward motion is ignored, the results become more and more dubious as this velocity builds up.

Gradual Propellant Release (Firestream):

If the damaged vehicle releases its propellants relatively slowly, or if it is traveling rapidly at the time of the abort, propellants will be spilled over a distance that is larger than the diameter of the fireball that these propellants could produce. It is under these conditions that the term "firestream" applies, and any model based upon a spherical fireball is then inappropriate.

The firestream is actually a very complex phenomenon and there exist virtually no experimental data on it. We approximate it here with a very simple model (as usual).

We assume that a cylindrical firestream is produced at the adiabatic flame temperature, implying the release of both the booster fuel and the oxidizer with good mixing. The cylindrical firestream is assumed to cool by radiation from the sides. Any additional cooling due to mixing with the ambient air is ignored. We also assume that the combustion gases quickly come to rest (or almost to rest) with respect to the ambient air, rather than continuing with the booster's forward velocity; a partial justification of this assumption is given in Appendix D.

In this model, the cooling rate is inversely proportional to the firestream diameter, which turns out to be proportional to $(\dot{W}/v_b)^{1/2}$, where $\dot{W}$ is the propellant release rate and v_b is the booster velocity. The velocity as a function of altitude was taken from the Saturn V mission profile as given in the SNAP 27 Safety Analysis Report⁽¹²⁾; other liquid-propellant vehicles will not differ greatly. The propellant release rate was

assumed to be 2.3×10^5 lb/sec, the mean value rather arbitrarily employed in Ref. 12.

For the reasons discussed earlier, released isotopic fuel was assumed to stay with the cooling gases and not follow the "hot spot" at the booster source. From the time-temperature history of these gases, the fraction vaporized was calculated using equations (2.3)-(2.5) exactly as before. These relations are based upon the assumption that the vaporization rate is diffusion-limited; for the smaller particles at the higher altitudes this ceases to be true and a correction was applied, using equation (A-5) in Appendix A.

Results are shown as a function of altitude for a variety of particle sizes in Fig. 14. For other values of $\dot{W}$, the fraction vaporized varies roughly as $(\dot{W})^{1/2}$.

Figure 14. Vaporization of $^{238}\text{PuO}_2$ fuels in aborts after lift-off as a function of altitude for particles of the size indicated by the numbers in parentheses.

VI. Recondensation

Vaporized radioisotopic fuel will not stay in the vapor state for long, especially if it is a relatively refractory material such as PuO_2 or one of the other actinide oxides. It is important to have some idea of the size and activity distribution of the radioactive particles produced when the vapor recondenses, since these parameters determine both subsequent atmospheric transport and radiobiological behavior. It is obviously impossible to come up with a precise analysis. Hence, we shall only go into enough detail to support certain general conclusions, which we state in advance:

1. Most of the activity will probably be attached to particles of inert material, especially smoke from the abort; relatively little will be in the form of particles of pure radioisotope compound. This conclusion could fail to hold if recondensation is from relatively concentrated vapor, say $\geq 10^4 \mu\text{Ci}/\text{m}^3$ (for ^{238}Pu).
2. Most of the activity will be attached to inactive particles a few microns in diameter or less, or it may possibly be in the form of submicron particles of radioisotope compound. In either case, it will be in the respirable size range and capable of traveling large distances down wind.
3. The amount of activity per particle will be highly variable, but there will not be many very highly active particles. Amounts of activity approaching or exceeding the maximum permissible body burden, or concentrations approaching or exceeding the maximum permissible, will probably represent large numbers of particles, rather than just a few intensely active particles. (This is not to say that other processes besides vaporization cannot yield particles of high individual activity!)

4. The above conclusions appear to hold for both liquid propellant and solid propellant aborts.

Liquid Propellant Systems

An important feature of the problem is that the vapor will be quite dilute (in chemical terms) when recondensation begins. To see this, let us consider a specific case, the SNAP-27 RTG in a Saturn V abort. The $^{238}\text{PuO}_2$ inventory is 3740 g or 4.5×10^4 Ci, of which we assume a fraction $\underline{f}$ is vaporized. The volume V_o of the Saturn V fireball is W/ρ_g where ρ_g is the density of the combustion gases, roughly 8.8×10^{-5} g/cm³ (Ref. 8). Hence, the concentration $\underline{C}$ is

$$C \approx 1.32 \times 10^{-10} f \frac{V_o}{V} \text{ g/cm}^3, \quad (6.1)$$

where the propellant weight $\underline{W}$ was taken to be 5.5×10^6 lb or 2.5×10^9 g, and $\underline{V}$ is the volume of gases through which the vapor is actually mixed.

In general, we must expect $\underline{V}$ to be less than V_o but not by many orders of magnitude, as the fireball's extreme turbulence should promote rapid mixing during the several seconds required for the temperature to fall to levels permitting recondensation. On the other hand, $\underline{f}$ is less than unity. Hence, the quantity $X = f V_o/V$ is not expected to be orders of magnitude different from unity unless $\underline{f}$ is exceedingly small (in which case the problem is of no great interest), and it is therefore convenient to express the problem in terms of this parameter. A value of $X = 1$ corresponds to the concentration that would result if the entire SNAP-27 fuel load were vaporized and uniformly distributed through a Saturn V fireball.

Assuming ideal gas behavior and $T \approx 2000^\circ\text{K}$, the partial pressure $\underline{p}$ of PuO_2 vapor corresponding to equation (6.1) is:

$$p = 8 \times 10^{-8} f V_o/V \quad (6.2)$$

If we assume $X \equiv f V_o/V \approx 1$, the saturated vapor pressure p_s equals p when $T \approx 2000^\circ\text{K}$, and about 15 to 20 seconds are required for the fireball to cool to this temperature (Fig. 1). Even if $X = 100$, the temperature must fall to 2300°K , which requires between 10 and 15 seconds. These relatively long times support the idea that the vapor will be mixed through a sizable fraction of the fireball before recondensation begins.

Depending upon the conditions, condensation can be either homogeneous (spontaneous nucleation of PuO_2 particles) or heterogeneous (condensation of PuO_2 upon preexisting particles of other materials which serve as nuclei). When the vapor cloud is dilute, homogeneous nucleation requires high values of the supersaturation ratio $S \equiv p/p_s$. Heterogeneous condensation can usually take place at lower values of S ; if there are sufficient suitable nuclei, the cloud will be depleted of vapor rapidly enough so that S will never build up to the high values required for homogeneous nucleation.

The competition between heterogeneous and homogeneous condensation can be expressed in semiquantitative terms. It is shown in Appendix E that heterogeneous condensation will deplete the cloud of vapor at a rate given roughly by

$$\frac{d \ln(p-p_s)}{dt} = -44.34R(T/M)^{\frac{1}{2}} \alpha n \bar{s} \quad (6.3)$$

where M is the vapor molecular weight, N is the number of particles serving as nuclei per cm^3 , $\bar{s}$ is the mean surface area per particle, and α is the "sticking coefficient." On the other hand, for PuO_2 , the supersaturation ratio S builds up at a rate which is approximately given by

$$\begin{aligned}
d \ln S / dt &= \frac{d \ln p}{dt} - d \ln p_s / dt \\
&= \frac{d \ln p}{dt} - \frac{6.74 \times 10^4}{T^2} \frac{dT}{dt}
\end{aligned} \tag{6.4}$$

Since homogeneous nucleation can occur only if $S \gg 1$, or $p \gg p_s$, we may replace $p - p_s$ in equation (6.3) with p and substitute into equation (6.4). If the result is negative or zero, S will never be able to build up to large values. Inserting numerical values for $T \approx 2000^\circ\text{K}$ gives, as the resulting criterion for having only heterogeneous condensation,

$$\alpha n \bar{s} > -1.7 \times 10^{-6} \frac{dT}{dt} \approx 1.7 \times 10^{-4}, \tag{6.5}$$

for a cooling rate dT/dt on the order of $-100^\circ\text{K}/\text{sec}$. If $\alpha n \bar{s} < -1.7 \times 10^{-6} dT/dt$, heterogeneous condensation will still occur, unless $n \bar{s} = 0$, but S will continue to rise until some homogeneous condensation will probably take place also. Values of $\alpha n \bar{s} \ll -1.7 \times 10^{-6} dT/dt$ would suggest that homogeneous nucleation will be the predominant process.

The problem, of course, lies in estimating $n \bar{s}$. If soot particles are present with a size distribution similar to that reported⁽¹⁵⁾ for "typical" polluted (smoky) air (count-median diameter = 0.075 microns, mass-median diameter = 0.5 microns), concentrations as low as 10^{-8} g/cm^3 will give $n \bar{s} \approx 10^{-3}$, which should promote heterogeneous condensation unless α turns out to be very low for PuO_2 on soot. For the case of the Saturn V, this amount corresponds to the conversion of less than 0.1% of the available carbon into soot. Visually, actual aborts are certainly smoky affairs, and the high luminosities observed for fireballs⁽⁸⁾ suggest the presence of large numbers of radiating particles. It is therefore tempting to conclude that condensation will be heterogeneous. On the other hand, thermochemical

calculations show that elemental carbon cannot exist in the fireball environment if chemical equilibrium is attained.⁽⁸⁾ Furthermore, the high-temperature heart of the fireball is not available to visual observation. The question is best regarded as open.

If the condensation is heterogeneous, the activity per particle will be roughly proportional to the particle's surface area for particles smaller than a few microns in diameter, as is shown in Appendix E. The mean activity is simply equal to the activity concentration of the vapor divided by the number concentration of particles. For the size distribution mentioned above and 10^{-8} g/cm³ of particles, the mean activity is about 1.08×10^{-15} X Ci/particle, where $\underline{X}$ is as defined previously. The corresponding particle activity distribution resulting for $X = 1$ is plotted in Fig. 15. The plot indicates that under these conditions, 95% of the total activity would be associated with particles having $d \leq 1$ micron and individual particle activities less than 0.1 pCi. For other conditions, the activity per particle is proportional to $X d^2 / n \bar{s}$. Though the values of these parameters used here are little more than guesses, it does seem likely that there will not be much activity in the form of particles with high individual activities; say greater than 1 pCi (10^{-12} curies). The range of activities would probably be considerably broader than Fig. 15 indicates, since the vapor concentration will vary considerably through the cloud while Fig. 15 was calculated for a single value of $\underline{p}$.

It has been suggested that relatively coarse particles (e.g., dirt) might be sucked up in the fireball and that these might mitigate the hazard by scavenging much of the vapor. In this connection it is worthwhile to consider the relative collection efficiency as a function of particle size,

Figure 15. Distribution of the total activity by particle size (left scale) and individual particle activity (right scale), calculated assuming heterogeneous condensation of $^{238}\text{PuO}_2$ vapor upon an inert aerosol. The curves have quantitative meeting only for the specific vapor concentrations and aerosol characteristics described in the text.

expressed on a weight basis. Figure 16 portrays the weight concentration required to give $-d\ell_p/dt > -d\ell_g/dt$ with $dT/dt = -100^\circ\text{K/sec}$. The density of the particle material was taken to be two and a temperature equal to 2000°K was assumed. The curvature results from the failure of equation (6.3) for the larger sizes, as discussed in Appendix A. The more accurate relation, equation (A-5), was used in this calculation. For particles with $d = .01$ cm, the mass of particles required would exceed 10% of the total mass of the propellants involved in the fireball, which seems improbable. Still higher concentrations would be needed to suppress homogeneous nucleation in fireballs smaller than the Saturn V's, since these would cool more rapidly. Likewise, higher concentrations would be necessary if $\alpha < 1$. Furthermore, particles such as soot with $d \approx 10^{-5}$ cm are over 1,000 times as efficient per unit mass in collecting the vapor (Fig. 16), and if they are present in any quantity most of the activity will be associated with them even if large quantities of coarse dust are also present. In general, it seems unrealistic to hope for much hazard reduction from this source.

If condensation nuclei of inert materials are scarce or absent, homogeneous condensation will result. Nucleation theory gives an estimate for the rate of formation $\underline{J}$ of nuclei (number $\text{cm}^{-3}\text{sec}^{-1}$)⁽¹⁵⁾:

$$\underline{J} = 5.5 \times 10^{31} \left(\frac{p}{T}\right)^2 \frac{\sqrt{\underline{Y}M}}{\rho} \exp[-17.56(M/\rho)^2(\underline{Y}/T)^3/(\ell_{nS})^2] \quad (6.6)$$

where $\underline{Y}$ and $\underline{\rho}$, are respectively, the surface tension and the density of the condensing material. The value of $\underline{Y}$ for PuO_2 was taken to be 525 dynes/cm.⁽⁵⁾ The theory also gives an estimate of the initial radius r_o of the resulting particles, which is

$$r_o = \frac{2\underline{Y}M}{RT_o \ell_{nS}} = 2.4 \times 10^{-8} \frac{\underline{Y}M}{\rho T_o \ell_{nS}} \quad (6.7)$$

Figure 16. Weight concentration of inactive particles (g/cm³) required to "scavenge" a dilute ²³⁸PuO₂ vapor at a given rate, illustrating the rapidly decreasing scavenging efficiency with increasing particle size (see text).

After nucleation, the particles grow by condensation of additional vapor at a rate given approximately by

$$\frac{dr}{dt} = 44.34\alpha (p/\rho)(M/T)^{\frac{1}{2}}. \quad (6.8)$$

Using equations (6.6) - (6.8) and the fireball time-temperature history, the processes of nucleation and subsequent particle growth could be followed as a function of time until the cloud is depleted of vapor; some more details are given in Appendix E.

The resulting distributions of the total activity with respect to individual particle activity are shown in Fig. 17 for initial vapor concentrations of 8×10^{-9} , 8×10^{-8} , and 8×10^{-6} atm ($X = 0.1, 1.0, \text{ and } 100$). For the first two values, the activity per particle is always extremely small. For the last value, the activities are much larger but still under 1 pCi. As noted earlier, values of $\underline{p}$ this large would require that the vapor be mixed through only a very small fraction of the total fireball volume. Coagulation can increase the activity per particle somewhat, but it is shown in Appendix E that the final activity is unlikely to exceed $\sim 2 \times 10^{-8}$ $\underline{p}$ curies/particle. Since $p \lesssim 10^{-5}$ seems almost certain, and $p \approx 10^{-7} - 10^{-8}$ seems more probable, the activity per particle is still expected to be less than 1 pCi.

Variations in $\underline{p}$ through the cloud will result in a considerably wider overall activity distribution than any one of the distributions shown in Fig. 17, since the activity per particle is rather strongly dependent upon the vapor density. This fact follows from equation (6.8) which shows that dr/dt is proportional to $\underline{p}$. For a given growth time the activity therefore varies as $\underline{p}^3$. The actual dependence of the activity upon $\underline{p}$ is slightly weaker, since the condensation is completed more rapidly as $\underline{p}$ increases

Figure 17. Distribution of total activity with respect to individual particle activities, assuming homogeneous recondensation of $^{238}\text{PuO}_2$ vapor in a Saturn V fireball. Initial vapor concentrations (partial pressures) assumed were as indicated. The width of each bar corresponds to a factor of two range in activity.

and so there is less time for particle growth.

The physical size of the PuO_2 particles resulting from homogeneous nucleation is given by $d = 0.24A^{1/3}$, where d is in microns, A is the activity in pCi and a specific activity of 12 Ci/g is assumed. The particles condensing from the lower vapor concentrations will be at most a few millimicrons in diameter. Particles this small tend to "condense" upon the somewhat larger inactive particles also present (either smoke or simply the natural aerosols) much as does vapor except that the process is substantially slower. It is unlikely to occur rapidly enough to permit much scavenging by any coarse dust present before the latter falls out; thus, such dust would have to be sufficiently abundant to suppress homogeneous nucleation in order to effectively reduce the hazard and, as suggested earlier, that seems unlikely. Particles as large as those formed at $p = 8 \times 10^{-6}$ could remain unattached to other aerosols for longer periods of time.

Solid Propellants

Most solid propellant formulations contain substantial amounts of aluminum. Hence, unlike the liquid-propellant case, one of the major combustion products is a solid, Al_2O_3 , and nuclei for heterogeneous condensation will be available in large quantity. On the other hand, the combustion gases cool much more rapidly.

From the flame temperature profile and the gas velocity (Section V), one may estimate the cooling rate to be of the order of 2×10^4 K/sec. Referring back to equation (6.5) then gives $\alpha n \bar{s} \gtrsim 0.034$ as the criterion for suppressing homogeneous nucleation. Crude estimates (Appendix E) suggest that $\alpha n \bar{s}$ may actually be as high as unity or even somewhat greater.

Consequently, it is probable that any vaporized radioisotope will condense upon these Al_2O_3 particles. Even if homogeneous nucleation also occurs, the resulting nuclei will be very small, $< 10^{-7}$ cm diameter according to equation (6.7), and they would have very little time for subsequent growth. Particles in this size range would soon attach themselves to the somewhat larger alumina particles giving almost the same result as if the vapor had condensed upon them in the first place.

The initial activity spectrum would be governed by the surface areas of the particles serving as condensation nuclei, as described for the liquid-propellant case. However, the concentration of alumina particles is expected to be very high initially, and considerable agglomeration may occur even in the brief time before the smoke plume is diluted with large amounts of air. The exact course of the process is impossible to predict. Even so, the crude analysis sketched in Appendix E suggests that the final particle sizes will still be too small to settle out rapidly, and most of the activity will again be associated with particles having activities $\lesssim 1$ pCi.

VII. Uncertainties and Approximations

Some of the uncertainties involved are inherent in the basic model or its method of application and apply similarly to all abort situations, and we shall consider these first. Others depend more strongly upon the individual abort category considered, and these will be taken up second.

General:

Uncertainties in the results calculated here are of two kinds. First, there are many uncertainties, some of them major, in the parameters used as input to the models employed. In addition, there were a number of mathematical approximations introduced in the models themselves.

One of the more important assumptions is that the particle size spectrum remains the same as at the time of release, except for vaporization. Actually, it is possible that the abort fire environments may rapidly degrade the metallic matrix of SSC type fuel forms, releasing the small ceramic particles. Indeed, the temperatures alone can exceed the melting point of the ceramic phase and approach or exceed the melting point of molybdenum. Furthermore, molybdenum, in particular, is oxidation-sensitive; thermochemical calculations based upon data from the JANAF tables¹⁶ indicates that reactions such as

can proceed to the right to an important extent. For example, the calculated equilibrium partial pressures of MoO_2 and MoO_3 total about 0.01 and 0.25 atm at 2500°K and 3000°K, respectively, if the $\text{H}_2/\text{H}_2\text{O}$ ratio is unity, and this estimate makes no allowance for the formation of species such as $(\text{MoO}_3)_3$,

which is highly exothermic¹⁷. The reaction rates are not known, but if they are controlled only by gas-phase diffusion, these partial pressures would imply molybdenum volatilization rates roughly 100 times as great as the PuO_2 vaporization rates. In a Saturn V fireball, for example, this would be sufficient to permit complete degradation of a cermet piece smaller than about one millimeter, and considerable attack on larger pieces. More oxidizing conditions would mean greater attack. Consequences of this attack differ for different abort situations, and will be considered separately in the appropriate subsections.

Clearly, experimental investigation of the response of cermet fuel forms to abort fires is highly desirable. Any such experiments should be designed to simulate both the thermochemical environment and also the rapid motion of the combustion gases across the sample.

In contrast to the situation with the metallic phase, thermochemical calculations suggest that abort fires are unlikely to provide a sufficiently reducing environment to seriously degrade the oxide phase; this problem was treated in Ref. 1.

There are many other uncertainties as to input parameters, although some of the material properties (thermal conductivity, specific heat) are known sufficiently accurately for present purposes. Results are also insensitive to errors in the emissivity except for the larger pieces of SSC fuel. More important is the effective vapor pressure of the SSC or PMC; measurements on molybdenum-coated specimens at 1600°C and 1700°C gave partial pressures of Pu species of 7.2×10^{-8} and 2.6×10^{-7} atm, respectively.⁽⁶⁾ In contrast, equation (2.5) gives 7.6×10^{-9} and 4.7×10^{-8} atm, respectively, at these temperatures. The data suggest that, at these

relatively low temperatures, the effective vapor pressure of the SSC is considerably higher than equation (2.5) implies, but also that it may be increasing less rapidly with temperature than predicted by equation (2.5). The effect may be due to small quantities of more volatile species, in which case the vapor pressure at the higher temperatures might be closer to what has been assumed here. Clearly, more data are needed.

Another highly uncertain parameter is the velocity $\underline{v}$ of fuel particles with respect to the fireball gases. Indeed, $\underline{v}$ may fluctuate rapidly due to fireball turbulence. However, results for the small particles are very insensitive to any reasonable variations in $\underline{v}$, and even the larger pieces show only a moderate (square root) dependence upon $\underline{v}$. Use of the terminal fall velocity here was based upon the idea that, if the velocity were initially much larger, the resulting drag forces upon the particle would be much greater than the particle's weight and it would be swiftly decelerated. In view of the lack of sensitivity to this parameter, especially for small particles, it seems unlikely that it will be a major source of error; see Appendix A for additional discussion.

There are also uncertainties about the properties of the combustion gases, the most important being the coefficient of diffusion for plutonium vapors within them. In general, this problem is probably less serious than some of those discussed above.

One of the significant mathematical approximations is the treatment of pieces of solid-solution cermet (SSC) and plutonia-molybdenum cermet (PMC) as if they were spherical. It does not cause major error if the size of the sphere is chosen to have the same surface/volume ratio as the particle. In the analysis of damaged SSC, the particle sizes quoted in

Table I were those used in equation (2.3). These irregular particles probably had surface areas that were actually somewhat larger than spheres of the same diameter and, hence, would probably vaporize somewhat more than calculated. Use of the infinite thermal conductivity approximation also underestimates the vaporization somewhat (the surface actually heats up slightly faster), but the effect is trivial except for the larger pieces which contribute very little anyway. Ignoring the temperature-dependence of certain material properties is believed to have slight effect. In the calculation for the intact disk, use of the temperature of the hottest part of the disk may result in a modest overestimate.

The neglect of heat transfer by conduction (and convection) from the combustion gases to the fuel particles might possibly be significant in certain cases. The approach used here was generally to assume radiative heating only for the large particles and instant heating for the smaller ones. In most practical cases, it appears that most of the vaporization will result from particles for which the latter assumption is very nearly true. For additional discussion, see Appendix B.

Liquid-Propellant Pad Aborts:

Some of the uncertainties involved in the Van Nice and Carpenter fire model⁷ have already been discussed. In brief, the model is probably on the severe side but it is believed that an actual abort could approach it fairly closely and there is not at present any justification for using a significantly less severe model. The generalized vaporization model developed in Section III involves whatever additional errors result from assuming that the Saturn V fireball model can be applied to other vehicles by scaling the time proportional to $W^{1/3}$.

It may be noted that the type of particle size spectrum found in impact tests with cermet fuel forms yields results that are not very sensitive to many of the uncertainties in vaporization rates. Much of the vaporization is due to "fines" which vaporize entirely unless the vaporization rate is very greatly reduced, while most of the remaining material is in the form of pieces large enough so that they contribute little unless the rate is greatly increased. Changes in rate less than a factor of ten were found to affect the final results by only a factor of two or less.

Unfortunately, this lack of sensitivity does not hold for the possibility that the fireball may disintegrate the metal matrix of a SSC or PMC type fuel, since this would radically alter the size distribution. If ceramic particles are released with the size distribution found after oxidation of the molybdenum matrix at lower temperatures (Column 3, Table 3.1), substantial vaporization will result even if several seconds are required to disintegrate the matrix, assuming a Saturn V fireball. For smaller fireballs, there is less time for both attack on the matrix and also subsequent vaporization, but the effect could still be considerable.

Solid Propellant Pad Abort:

The greatest single uncertainty probably lies in modeling the fire, especially with respect to estimating the all-important maximum temperature to be expected. Use of the adiabatic flame temperature appears to be a much poorer approximation in this case than it was for liquid propellants. This is not surprising, since much of the calculated energy release derives from aluminum oxidation which is a heterogeneous reaction and which is experimentally observed to be incomplete. In addition, the time available for complete reaction is much less. The adiabatic fire

model of Section IV therefore is almost certainly an overstatement, while the semi-empirical model could be in error by a sizable factor in either direction.

When cermet fuel forms are involved, matrix disintegration is again a distinct possibility for those particles too large to blow off. However, in this case, any ceramic particles freed will be in the blow-off size range. Thus, the result of matrix disintegration in a solid propellant fire may be the production of large amounts of "fines" having $d < 100$ microns with only a moderate amount of true vaporization.

Another important problem is the question of whether pieces of cermet-type fuel will actually be exposed to the hottest part of the fire assuming they are too large to blow off. Though it may be quite close, the hottest region cannot be right at the surface of the burning propellant because the solid itself cannot be extremely hot. Hence, pieces of fuel resting upon the surface might see significantly lower temperatures than has been assumed. A reduction of 200° from the 2515°K temperature assumed in the semi-empirical model would reduce vaporization by a factor of ten.

The assumption that particles are at the fire temperature when released could result in an overestimate of the vaporization for the larger particles which are still within the blow-off size range. However, only slight vaporization is predicted for these anyway, so this error is not very important.

Certain approximations (spherical particles, Stoke's law, etc.; see Appendix D) were introduced in order to integrate the equations of motion for those particles which blow off. These are less important, however, than the fact that the whole model is undoubtedly much too neat, without the

complex variability and unpredictability which are expected to characterize any actual abort fire. Nonetheless, the basic idea that small particles will be quickly blown out of a solid propellant fire is very probably correct.

Altitude Aborts:

The firestream model is one of the major sources of uncertainty in the case of altitude aborts. Mixing with air probably should not be neglected. The model assumes good mixing of the propellants which were implicitly taken to be released with the same mixing ratio as they are present in the tankage. All effects due to vehicle motion, including the high Mach numbers attained at high altitudes, were neglected.

In addition, it was assumed that any radioisotopic fuel released would be ejected directly into the hottest part of the firestream. This would presumably require the vehicle to reverse orientation and fly backwards with the payload bathed in the firestream. While such could happen, the probability may not be very high. Indeed, it seems perfectly possible for released fuel to never enter the firestream at all. The situation differs from the pad abort case, where the fireball completely engulfs the vehicle and any released fuel will very likely be exposed to the fireball environment.

On the other hand, the temperature response of released particles was calculated here assuming only radiative heating for particles of all sizes. For particles less than 100 microns in diameter, the thermal lag effects are still small; for much larger pieces, the assumption of radiative heating is essentially correct. For particles of the order of 200 microns in diameter, ignoring conductive heating may underestimate the vaporization,

especially at the higher altitudes. The effect is somewhat larger here than in the pad abort case because the firestream cools more rapidly and a significant thermal lag is predicted down to smaller particle sizes if only radiative heating is assumed.

Attack upon the metallic matrix of cermet fuel forms would have a qualitatively similar effect as in the pad abort case. The time over which such attack can occur is less, however.

In general, the uncertainties are greater for an altitude abort. Nonetheless, it seems safe to conclude that an abort at high altitude will result in no more, and probably less, vaporization than will a pad abort.

VIII. Conclusions

The rate of vaporization of radioisotopic fuels in abort fires is found to be a strong function of particle size. For small particles (say, less than a few hundred microns in diameter) vaporization can be substantial in the pad abort of any liquid-propellant vehicle, though this will not be true for the case of small vehicles unless the radioisotope is released promptly upon fireball initiation. Vaporization may be less for fuel forms such as SSC and PMC, but it will not be negligible unless the cermet disks retain complete integrity, which is improbable in any abort sufficiently violent to result in containment failure and fuel release. Vaporization of cermets could be greatly enhanced if the metallic matrix is rapidly disintegrated by the fireball's thermochemical environment (thermodynamic data suggest that this is a possibility). Uncertainties in the analysis are greater for altitude aborts, but in general these are expected to result in less vaporization than pad aborts.

The model developed for solid-propellant fires takes into account the fact that small particles are very likely to be quickly blown out of the fire, while larger pieces may remain. As a result, pieces of cermet fuels released to a solid-propellant fire may actually undergo a greater percentage of vaporization than more finely divided fuel forms such as microspheres. For the latter type of fuel, solid-propellant fires probably present less vaporization hazard than do liquid-propellant fires. Altitude aborts of solid-propellant vehicles appear to involve relatively little vaporization hazard.

After the vapor recondenses, most of the activity will probably be associated with relatively small particles, a few microns in diameter or

less; and hence will be in the respirable size range and will be capable of long-distance atmospheric transport. The activity per individual particle will usually be sufficiently small so that biologically hazardous quantities will represent large numbers of particles, rather than just a few highly active particles.

The many uncertainties involved in the analyses must be stressed. Some of these uncertainties result from the fact that an abort is, by its very nature, a violent, unprogrammed event largely governed by parameters which can vary essentially at random over a wide range. The maximum temperature obtainable is an important source of uncertainty, especially for solid-propellant fires. The effective vapor pressure of fuel forms other than pure $^{238}\text{PuO}_2$ has not been adequately determined at high temperatures. For cermet fuel forms, the response of the metallic components to the thermochemical environment of the fire is essentially unknown, but it could seriously affect the vaporization behavior. Experimental investigations are recommended.

9

APPENDIX A

FUNDAMENTAL RELATIONS FOR VAPORIZATION
AND CONDENSATION

Appendix A

Fundamental Relations for Vaporization and Condensation

Vaporization into a gaseous medium which is at rest with respect to the particle will be considered first. The enhancement which can result from the relative motion of the gases and the particle (convection) will be briefly discussed at the end of this Appendix.

Let us assume that a spherical particle of radius a is suspended in a gaseous medium containing, at large distances, a concentration C_∞ of the particle's vapor. Let p_s be the saturated vapor pressure of the material and p_b be the partial pressure of vapor in the boundary layer at the surface of the particle; we do not assume that $p_b = p_s$. The net rate at which the particle loses molecules is given by the relation¹⁸

$$n(\text{molecules cm}^{-2}\text{sec}^{-1}) = \alpha(p_s - p_b)/(2\pi mkT)^{1/2}, \quad (\text{A-1})$$

where m is the molecular mass, k is the Boltzmann constant, T is the absolute temperature, and α is a "sticking coefficient." If $p_b = 0$, equation (A-1) becomes the expression for the vaporization rate in a vacuum.¹⁸ Converting to more convenient units, we have a rate of weight loss per unit area, w/A , of

$$w/A(\text{g cm}^{-2}\text{sec}^{-1}) = 44.34\alpha(p_s - p_b)(M/T)^{1/2} \quad (\text{A-2})$$

where the p 's are now in atmospheres and M is the molecular weight.

This rate must be equal to the rate J_b at which vapor diffuses away from the boundary layer, given by

$$J_b = -D \frac{dC}{dr} \bigg|_{r=a}$$

where $\underline{D}$ is the diffusion coefficient and the concentration $\underline{C}$ is related to the partial pressure of vapor $\underline{p}$ by $C = Mp/RT$, where $\underline{R}$ is the gas constant. Since the vapor is very dilute, we may assume a quasi-steady state exists near the particle; that is, the amount of vapor diffusing through a spherical shell of radius $\underline{r}$ is independent of $\underline{r}$, and therefore $4\pi r^2 \cdot J(r) = 4\pi a^2 J_b$, or $J(r) = J_b a^2/r^2$. Hence,

$$J(r) = J_b a^2/r^2 = -D \frac{dC}{dr},$$

$$C = \frac{J_b a^2}{Dr} + k.$$

At $r = \infty$, $C = C_\infty$; hence, $k = C_\infty$. At $r = a$, $C = C_b$; hence, J_b is given by

$$J_b = \frac{2D}{d}(C_b - C_\infty) = \frac{2D}{d} \frac{M}{RT}(p_b - p_\infty) = 2.437 \times 10^{-2} \frac{D}{d} \frac{M}{T}(p_b - p_\infty). \quad (A-4)$$

where $d = 2a$ is the diameter of the particle.

We next eliminate p_b between equations (A-2) and (A-4), since it is not known (p_s and p_∞ are taken as given). For convenience, we set $K_1 = 44.34\alpha(M/T)^{1/2}$, and $K_2 = 2.437 \times 10^{-2} DM/Td$. We then obtain,

$$w/A = \frac{K_1 K_2}{K_1 + K_2} (p_s - p_\infty). \quad (A-5)$$

We note that if $K_2 \gg K_1$, equation (A-5) reduces to (A-2) with $p_b = p_\infty$ and the vacuum vaporization relation controls the vaporization, with the rate per gram simply proportional to the surface/mass ratio, which varies as $1/d$. On the other hand, if $K_2 \ll K_1$, equation (A-5) reduces to (A-4) with $p_b = p_s$. Diffusion away from the boundary layer then controls the vaporization, and the rate per gram is proportional to (surface/mass)/ d , or $1/d^2$.

To estimate the dividing line between the two regimes, we set $T = 2500^\circ\text{K}$, $M = 270$ (for $^{238}\text{PuO}_2$). Then $K_1 = 14.57\alpha$ and $K_2 = 2.63 \times 10^{-3} D/d$. In Ref. 1, $\underline{D}$ was estimated to be about $1.47 \times 10^{-5} T^{1.62}$ for $T > 1000^\circ\text{K}$ and one atmosphere ambient pressure; hence, at 2500°K , $D \approx 4.7 \text{ cm}^2/\text{sec}$. K_1 and K_2 are then equal for $d \approx 8.5 \times 10^{-4}/\alpha \text{ cm}$, or 8.5 microns if $\alpha = 1$. The value of $\underline{\alpha}$ is not known for PuO_2 , but there is some evidence that it does not differ greatly from unity³, and $\alpha = 1$ has been assumed for all numerical calculations in this report.

In the case of the vaporization calculations, we were generally interested in particles larger than 10 microns, and diffusion-control dominates. Consequently, all sea-level vaporization calculations were based upon equation (A-4), modified for convective enhancement⁽¹⁾, and with $p_\infty = 0$ (since $p_\infty \ll p_s$ in this case). Some slight overestimate results for the smallest particles. At higher altitudes, this overestimate increases due to the increase in $\underline{D}$, and a correction based upon equation (A-5) was applied.

Nothing in the preceding derivations depended upon any assumption that $p_s > p_\infty$. If $p_\infty > p_s$ the sign of the mass flow is simply reversed; i.e., we have recondensation occurring governed by exactly the same relations. In the case of recondensation, however, we are dealing either with the extremely small particles formed by homogeneous nucleation or with recondensation on inactive particles (soot, etc.) which may be somewhat larger but are still of the order of 10^{-4} cm or less in diameter. Hence, the rate will be governed by the vacuum vaporization relation, and equation (A-2) was usually employed in treating the recondensation problem, with $p_b = p_\infty$. Equation (A-5) was used in the one case where recondensation was considered as a function of particle size over a wide range (Fig. 16).

In the approach utilized here, the vapor is treated as flowing against two "resistances" in series, with $p_s - p_\infty$ being the driving force or "voltage". One of the two "resistances" is the vacuum vaporization limit (the rate molecules can strike or leave the surface) while the other is set by diffusion of vapor away from the particle. The K's in equation (A-5) are analogous to conductances. The treatment of the diffusion term becomes inadequate for very small particles because all effects due to the finite molecular mean free path λ have been ignored. However, that is of small consequence here, since $\lambda \lesssim 10^{-4}$ cm (at sea level) for all cases of interest and the vacuum vaporization relation controls the rate for particles in this size range. The same statement holds at high altitudes also, since λ and D both vary inversely with the atmospheric pressure.

It was assumed in the preceding analysis that the vaporizing particle was at rest with respect to the surrounding gases. If it is actually in motion, the basic vacuum vaporization limiting rate is not affected but the vaporization rate in the diffusion-controlled regime can be significantly enhanced. This convective enhancement was taken into account by using the semi-empirical mass-transfer relation⁽²⁾,

$$w/A = 2 \frac{D}{d} \left[1 + 0.276 \text{Re}_d^{1/2} \text{Sc}^{1/3} \right] \frac{p_s M}{RT} \quad (\text{A-6})$$

as was mentioned in Section II. This relation is based on the assumption of laminar flow about the particle. In the present work, the Reynold's numbers were always below 2×10^4 even for the largest particles, while the critical value for the onset of turbulent flow is greater than 10^5 .

In so far as the "local" flow around the particle is concerned, then, the assumption of laminar flow should be valid. However, it is also

assumed in equation (A-6) that the general motion (if any) of the main body of gas is laminar, while in the case of a fireball it is actually extremely turbulent. Since the scale of the turbulence is much larger than the scale of the flow about the particle, one may think of the latter flow as being laminar but with a variable velocity $\underline{v}$ which is at least partly governed by the larger-scale turbulent motions. Unfortunately, there is no realistic way to estimate this velocity.

In the present work, $\underline{v}$ was simply set equal to the terminal fall velocity v_t , the argument being that, if $v \gg v_t$, the drag force on the particle will greatly exceed the weight of the particle, and it will be quickly decelerated. Undoubtably this assumption is a crude one. Fortunately, the results are rather insensitive to anything but a very drastic failure of the assumption. For example, a (spherical) particle with $d = 0.015$ cm, $\rho = 10.5$ g/cm³ will have $v_t \approx 155$ cm/sec. If we assume $v = 10v_t = 1.55 \times 10^3$ cm, the resulting drag force is equivalent to about 13.5 times the acceleration of gravity, or 1.3×10^4 cm/sec², which will quickly reduce the excess velocity unless the surrounding gases are also continually being subjected to accelerations this large. Furthermore even this large a velocity would only increase the vaporization rate by about 33% over the value given by $v = v_t$, according to equation (A-6).

For larger particles, the dependence of the vaporization rate upon $\underline{v}$ is somewhat greater, but for these, the terminal fall velocities are already of the order of the turbulent gas velocities. For example, at a temperature of about 2700°C, $v_t = 2.3 \times 10^3$ cm/sec for $d = 0.1$ cm and $v_t = 1.6 \times 10^4$ cm/sec if $d = 1.0$ cm, while the observed gas velocities⁽⁸⁾ are of the order of 200 ft/sec or 6.1×10^3 cm/sec. Since the dependence of the

vaporization rate upon $\underline{v}$ is never stronger than $v^{1/2}$, it again seems unlikely that effects due to turbulence can give any great increases in the vaporization rate over the values calculated for $v = v_t$.

APPENDIX B

NOTE ON HEAT TRANSFER MECHANISMS

Appendix B

Note on Heat Transfer Mechanisms

If radioisotopic fuel is released into an abort fire, the pieces of fuel may initially be at temperatures much lower than the fire temperature, and will not vaporize significantly until they have been heated up. For very small particles, the time required is small compared with the duration of a liquid-propellant fireball, but it becomes significant for pieces of fuel a few hundred microns or more in diameter, especially for the smaller fireballs. Consequently, an attempt was made to take this "thermal lag" into account in the present work.

It was initially assumed that heat transfer would be primarily by radiation, since many other analyses of the heat-transfer problem in fireballs had shown radiation to dominate overwhelmingly at the high temperature involved^{7,8,12}. All calculations of thermal response were therefore made assuming only radiative transfer and neglecting conduction from the hot gases. However, more careful consideration has shown that this conclusion must be qualified; in particular, heat conduction from the gases would appear to dominate the process for sufficiently small particles. ("Conduction", in this context, should be understood to include any enhancement due to convection.)

The mathematics of heat conduction are essentially identical to the mathematics of diffusion. Furthermore, the heat capacity per unit volume of the solid is about 4×10^4 times as great as that of the surrounding gases at $\sim 2500^\circ\text{K}$. Hence, we can again make the assumption that a quasi-steady state for the heat flow will exist just as we did for the diffusion of vapor in Appendix A, though perhaps not with quite the same degree of

validity. We may then write, in analogy with equation (A-4),

$$\dot{\Phi}_c = \frac{2K}{d} \Delta T, \quad (B-1)$$

where $\dot{\Phi}_c$ is the heat flux per unit area to the particle surface due to conduction, K is the thermal conductivity of the gases, and ΔT is the temperature difference between the particle and the fireball.

The thermal conductivity for a pure gas may be estimated from the relation⁽¹⁹⁾

$$K \approx \frac{1}{4} (9\nu - 5) \eta C_v / M \quad (B-2)$$

where $\nu = C_p / C_v$ is the ratio of specific heats, C_v is the molar specific heat at constant volume, and η the viscosity. Values of C_v for the gases of interest (see below) were taken from the JANAF tables¹⁶ while η was estimated from the approximate relation¹⁹

$$\eta \approx aT^n / (1 + C/T), \quad (B-3)$$

using values of the parameters n and c taken from Ref. 19.

The net heat flux $\dot{\Phi}_r$ to the particle surface per unit area due to radiation is given simply by

$$\dot{\Phi}_r = \sigma \epsilon (T_f^4 - T^4) \approx 4\sigma \epsilon T^3 \Delta T \text{ if } \Delta T \ll T, \quad (B-4)$$

where σ is the Stephen-Boltzmann constant and ϵ is the emissivity of the particle. It is instructive to compare this value with the heat transfer by conduction. For the present purposes, it will be sufficient to assume all but one of the major fireball constituents have a conductivity similar to that of one of the principle constituents, carbon monoxide. The clear

exception will be hydrogen, which has a much higher conductivity. Thermochemical calculations suggest that, for some solid propellant combinations, the hydrogen content of the combustion products may be as high as 30%. For mixtures, the thermal conductivity is approximately given by the weighted average, $K \approx \sum K_i X_i$, where K_i and X_i are, respectively, the conductivities and the mole fractions of the individual components in the mixture (this relation is not very precise).

In Table B-1 below we give the net heat flux/ ΔT for radiation (assuming $\epsilon = 0.65$ and $\Delta T \ll T$) and for conduction, assuming 0% and 30% hydrogen. Note that the conductive transfer varies as $1/d$, while the radiative transfer is independent of d .

Table B-1
Heat Transfer Rates ($\text{cal deg}^{-1} \text{cm}^{-2} \text{sec}^{-1}$)

T (°K)	Radiation	Conduction	
		0% H ₂	30% H ₂
2000	0.0282	.00047/d	.00130/d
2500	0.0551	.00053/d	.00154/d
3000	0.0951	.00063/d	.00178/d

These numbers are, of course, rough estimates, and hold only for $|\Delta T| \ll T$. For large ΔT , the temperature-dependence of K must be taken into account which means that equation (B-1) no longer holds, and also the first form of equation (B-4) must be used for the radiation case.

From Table B-1, it is clear that conduction is the dominant heat transfer mode for sufficiently low T and small d . The importance of the question of the hydrogen content is also obvious. With $d \geq 0.02$ cm and hydrogen

absent, radiation would appear to dominate for $T \geq 2000^\circ\text{K}$; with as much as 30% hydrogen, conduction dominates almost up to 3000°K for $d = 0.02$.

Whether this much hydrogen will be present is not clear; the presence of unburned aluminum after solid propellant burn tests (which shows thermochemical equilibrium was not obtained) suggests that more oxygen may be available for combination with hydrogen than is implied by the thermochemical calculations. Liquid-propellant combinations are generally less oxygen-deficient than the solids, but some smaller amount of free hydrogen is still expected to be present.

In the actual cases of interest, the approximations based upon $\Delta T \ll T$ are not generally useful. Considering the importance of such uncertainties as the amount of free hydrogen present, it seems questionable as to whether detailed treatment of all effects involved is justified; in any case, it was not permitted by the time and resources available to the present program.

In liquid-propellant aborts, radiative heating only was included for $d > 0.02$ cm; for smaller particles, thermal lag was simply ignored (i.e., instantaneous heating assumed). Vaporization is underestimated in the first case and overestimated in the second; in both cases, the error is probably not very important because, even assuming only radiative heating, the thermal lag effects are not large except for considerably larger particles, for which the assumption is probably fairly valid.

In the solid propellant blow-off model, particles as small as $\underline{d} = 10^{-3}$ cm are predicted to show significant thermal lag if only radiative heating is considered; however, a glance at Table B-1 suggests that conductive heating will actually be much the greater for particles of this size. Hence, it was simply assumed that all particles were at the fire temperature when

released. This assumption may overestimate the vaporization, but not greatly for the smallest particles, which are the ones that contribute the most. Furthermore, the assumption may actually be more or less correct if fuel release is due to capsule thermal failure. Relative motion of the particle and the surrounding gases (convection) can enhance the transfer of heat by conduction; indeed, the enhancement factor is essentially the same as for the enhancement of the mass transfer by diffusion. Once again, the effect becomes substantial only for the larger particles for which radiation will probably still be the dominant heating mechanism and which, in any case, undergo relatively little vaporization.

APPENDIX C

"BLOW-OFF" MODEL FOR VAPORIZATION IN SOLID PROPELLANT FIRES

Appendix C

"Blow-Off" Model for Vaporization in Solid Propellant Fires.

The propellant UTP-3001 nominally yields upon burning about 0.0348 moles of gaseous products per gram⁽²⁰⁾. With a burning rate of 0.06"/sec⁽¹¹⁾ and a density of 1.8 grams/cm³, the net rate of gas generation is .0095 moles cm⁻²sec⁻¹ which corresponds to 1970 cm³ of gas/cm²sec at 2515°K. Since this volume of gas must flow away from each square centimeter of the burning surface every second, it must be equal to v_g , the net velocity of the gas away from the surface.* Turbulence can increase the absolute magnitude of the instantaneous velocity.

If a particle is held in a gas flowing with a velocity $\underline{v}$ relative to the particle, it will experience a drag force

$$F = \frac{1}{2} C_D \rho_g v^2 A = \frac{1}{8} \pi d^2 \rho_g v^2 C_D, \quad (C-1)$$

where C_D is the drag coefficient, A is the crosssectional area presented by the particle to the flow, and we approximate the particle as a sphere; hence $A = \pi d^2/4$. In general, C_D is a complicated function of the Reynolds number $Re_d = vd/\nu$ which has been determined by largely empirical means⁽²¹⁾. If the gas flow is in the upward direction, the net force on the particle is obtained by subtracting off the particle's weight, $\frac{1}{6} \pi g d^3$, where $\underline{g}$ is the acceleration of gravity (we neglect the trivial effect of bouyancy upon the particle). The rate of change of the particle's velocity v_p is then found by dividing this force by the mass:

*The actual calculations were performed using $v_g = 2129$ cm/sec due to the use of slightly different input assumptions in earlier calculations. The consequences of this difference in v_g are trivial compared with the general level of uncertainty.

$$\frac{dv_p}{dt} = \frac{3}{4} \frac{\rho_g}{\rho} v^2 C_D - g \quad (C-2)$$

By setting this equal to zero and solving, we may obtain the terminal fall velocity, v_t :

$$v_t = \left(\frac{4}{3} \frac{\rho_g d}{\rho C_D} \right)^{\frac{1}{2}} \quad (C-3)$$

Since C_D is a function of $\underline{v}$, equation (C-3) can be solved directly for v_t only in certain special cases, but iterative techniques may always be used whenever C_D is known as a function of $\underline{Re}$.

Using equation (C-3), v_t was calculated as a function of particle diameter with $\rho = 10.5$; the results are the terminal velocity values used throughout this report. If C_D did not depend upon $\underline{v}$, one could use equation (C-3) to substitute for C_D in equation (C-2) to obtain dv_p/dt in terms of v_t . It turns out to be convenient to write $C_D(v) = C_D(v_t) \cdot C_D(v)/C_D(v_t)$ and carry out the substitution anyway, obtaining

$$\frac{dv_p}{dt} = g \left(\frac{v^2}{v_t^2} \frac{C_D(v)}{C_D(v_t)} - 1 \right) \quad (C-4)$$

For sufficiently small particles, Stoke's law applies and C_D varies as $1/v$; this law holds to within a few percent for $Re \leq 1$. For larger values of Re , C_D decreases more slowly with increasing $\underline{v}$, but the deviation from proportionality to $1/v$ is gradual. For $Re = 20$, Stoke's law underestimates C_D by slightly more than a factor of two; this value of Re corresponds roughly to $v = v_g \approx 2 \times 10^3$ cm/sec, $d = 0.05$ cm and is about as high a value as will be of interest to us here.

In analyzing the blow-off of particles, v_t was calculated from

equation (C-3), using the correct value of C_D (Ref. 21) and iterating where necessary. Then the assumption that C_D varies as $1/v$ was made in equation (C-4), giving

$$\frac{dv_p}{dt} = g\left(\frac{v}{v_t} - 1\right) = g\left(\frac{v_g - v_p - v_t}{v_t}\right), \quad (C-5)$$

since $v = v_g - v_p$. We are only interested in cases where $v \geq v_t$, i.e., when the particles will blow off, and our procedure therefore underestimates C_D when the approximation is not valid; hence, the time required for blow-off, and therefore the vaporization, may be somewhat overestimated for the larger particles. Another moderate overestimate comes about because v_t was calculated assuming a spherical shape. Actually, the particles will be irregular (unless we are dealing with microspheres) and v_t will be somewhat smaller than for a sphere of the same cross sectional area¹⁵.

To continue, we next assume that the particles are initially at rest and integrate equation (C-5), obtaining

$$v_p = (v_g - v_t)(1 - e^{-gt/v_t}) \quad (C-6)$$

Assuming that the particles are initially resting on the propellant surface, we integrate again to obtain the height H above the surface as a function of time:

$$H(t) = (v_g - v_t)\left[t - \frac{v_t}{g}(1 - e^{-gt/v_t})\right] \quad (C-7)$$

Given $H(t)$ and the flame temperature profiles of Fig. 9, the temperature of the particle was calculated as a function of time and the vaporization then estimated essentially as in other cases. The convective enhancement factor in equation (2.3) was evaluated with $v = v_g$ at all times instead of the more

nearly correct value $v_g - v_p(t)$, which is continually changing as a function of time; the error resulting from this simplification is small.

Finally, particles large enough so that $v_t > v_g$, with v_t calculated from equation (C-3), were assumed to remain in the fire until either they vaporized entirely or until the fire burned out.

APPENDIX D
ALTITUDE ABORTS

Appendix D

Altitude Aborts

Only liquid-propellant vehicles are considered here for reasons mentioned in Section V. The altitude abort problem was approached in two different ways. In the first, the pad abort fireball model was simply extrapolated to higher altitudes by taking into account certain factors which depend upon the ambient pressure. Such a model might apply to an abort in which a violent explosion demolishes the tankage and abruptly releases all propellants. In the second, a very simple model is offered for a cylindrical firestream, which may be more appropriate to an abort in which propellant is released more slowly, or when the forward velocity is high. In neither case are effects due to this forward velocity meaningfully taken into account.

Abrupt Release:

The adiabatic flame temperature is plotted as a function of atmospheric pressure $\underline{P}$ in Fig. D-1a, as calculated by means of the ACE computer code^{8,14}, using the Saturn V propellant compositions. The decline in temperature with decreasing $\underline{P}$ reduces the vapor pressure p_g , but this is compensated for by the increase in the diffusion coefficient $\underline{D}$. The vaporization rate $\underline{Y}$ is proportional to the product $\underline{D}p_g$, other things being equal; hence, we can write

$$\frac{d \ln Y}{dP} = \frac{d \ln D}{dP} + \frac{d \ln (p_g)}{dP}, \quad (D-1)$$

where $\underline{P}$ is the ambient pressure. The diffusion coefficient varies essentially as P^{-1} . From the vapor pressure equation (2.5) we may write

Figure D-1. Adiabatic flame temperature (a) and combustion-gas heat capacity at constant pressure (b) as a function of ambient pressure for the Saturn V propellant combination. The heat capacity is calculated at the corresponding adiabatic flame temperature for each pressure, and it is assumed that chemical equilibrium is continually maintained as the temperature changes.

$$\frac{d \ln(p_s)}{dP} = \frac{67374}{T^2} \frac{dT}{dP} \quad (D-2)$$

The curve in Fig. D-1a can be fit to within a few degrees by

$$T \approx T_o + 108 \ln P \quad (D-3)$$

Inserting equations (D-2) and (D-3) into equation (D-1) and evaluating at $T = 2850^\circ\text{K}$ as representative of the range of interest in Fig. D-1a, we obtain

$$\frac{d \ln Y}{dP} \approx -1/P + .896/P = -.104/P \quad (D-4)$$

$$Y \approx Y_o P^{-0.104}$$

Thus, the net vaporization rate shows only a very weak dependence upon $\underline{P}$, corresponding to a 40% change for a factor of a hundred change in $\underline{P}$.

The above applies only to the initial moment, when the fireball is at the adiabatic flame temperature. The fireball then proceeds to cool at a rate

$$\frac{dT}{dt} = - \frac{\pi d^2 \sigma \epsilon T^4}{\rho_g c_p \cdot \frac{\pi d^3}{6}} = \frac{6 \sigma \epsilon T^4}{\rho_g c_p d} \quad (D-5)$$

Here, the density of the gases ρ_g varies as P/T , the diameter $\underline{d}$ varies as $(T/P)^{1/3}$, assuming the total mass of the fireball is constant; hence,

$$\frac{dT}{dt} \propto P^{-2/3} T^{14/3} / c_p \quad (D-6)$$

If the curve of Fig. 1a is fitted with a power law, one finds that $T = T_o P^{.0388}$ gives a fit to within less than 10°. The constant pressure heat capacity c_p depends strongly upon both pressure and temperature;

Fig. 1b gives the values calculated⁽¹⁴⁾ as a function of pressure at the corresponding adiabatic flame temperature. The curve may be fitted to within a few percent by $c_p \approx c_p^0 P^{-0.150}$. Inserting these relations into equations (D-6), one obtains

$$\frac{dT}{dt} \propto P^{-.34} \quad (D-7)$$

Thus the initial cooling rate also shows only a rather weak dependence upon the pressure.

The initial vaporization rate is therefore seen to be almost altitude-independent, while the initial rate of cooling increases with altitude (decreasing P), but only rather slowly. In any altitude about where a spherical fire model is felt to be appropriate at all, it appears that the vaporization will not differ greatly from that of the pad abort case.

Cylindrical Firestream Model:

In many cases, perhaps most, the vehicle may release propellants over a time during which it travels a distance that is large compared with the diameter of any possible fireball. Any spherical model is then inappropriate. We assume, instead, a cylindrical firestream, at the adiabatic flame temperature near the point of propellant release (the booster vehicle) with the gases subsequently cooling from the sides by radiation only, without additional cooling due to air admixture.

Proceeding as in equation (D-5), we obtain

$$\frac{dT}{dt} = - \frac{\pi d \sigma \epsilon T^4}{\rho_{g,p} c_p \cdot \frac{1}{4} \pi d^2} = - \frac{4 \sigma \epsilon T^4}{\rho_{g,p} d}, \quad (D-8)$$

since πd and $\frac{1}{4} \pi d^2$ are, respectively, the surface area and the volume per unit length. The firestream diameter d may be estimated by noting that the

spillage of more propellant is adding combustion gases of volume $\dot{W}/\rho_g$ per unit time, where $\dot{W}$ is the propellant release rate, and this must be equal to $\frac{1}{4}\pi d^2 v_b$, where v_b is the booster velocity. Solving for d and substituting in equation (D-8) gives

$$\frac{dT}{dt} = - \frac{2\pi\epsilon}{c_p} \left(\frac{\pi v_b}{\rho_g \dot{W}} \right)^{1/2} T^4 \quad (D-9)$$

Numerical calculations reported here were performed with $\dot{W} = 2.3 \times 10^5$ lb/sec = 1.04×10^8 g/sec, the value somewhat arbitrarily assumed in Ref. 12 for a Saturn V abort. Since the cooling rate varies as $1/\dot{W}^{1/2}$, this model predicts the vaporization will be proportional to $\dot{W}^{1/2}$ for particles whose temperature follows the fire temperature without a significant lag. The value of v_b as a function of altitude was taken from the Saturn V mission profile¹². The density of the gases was assumed to be proportional to P . The additional dependence upon T was ignored because of the relatively narrow range of temperature over which vaporization will contribute significantly. The heat capacity c_p was calculated⁽¹⁴⁾ assuming chemical equilibrium would be maintained as the temperature changed; the resulting values are shown as a function of temperature for several different pressures in Fig. D-2. The effective heat capacity rises rather sharply with increasing temperature and decreasing pressure because of the increasing chemical dissociation of the reaction products.

Once the time-temperature history of the combustion gases was obtained, the vaporization could be calculated in essentially the same manner as with the other abort situations, except for one modification. At high altitudes, the diffusion coefficient becomes sufficiently large so that diffusion of vapor away from the particle is no longer the limiting factor on the

Figure D-2. Heat capacity of the Saturn V combustion gases as a function of temperature, calculated for the indicated values of the ambient pressure. Chemical equilibrium is assumed to be maintained as the temperature changes.

vaporization rate, especially for the smaller sizes. Consequently, equation (A-5) of Appendix A was employed for these calculations, in order to take into account the fact that the rate cannot exceed the fundamental vacuum vaporization rate.

It has been implicitly assumed here that the combustion gases very rapidly lose the booster's forward velocity. As a plausibility argument, we may note that, due to the temperature difference, these gases possess less than a tenth the density (and therefore the inertia) of the air they displace, somewhat as if a balloon were to be fired into water. Another approach is to compare the momentum of the propellants released per unit time with the drag on the front surface of an object having the cross section estimated for the firestream; this drag turns out to be about an order of magnitude greater than the momentum, and since force equals the time rate of change of momentum, most of this momentum should be lost in a fraction of a second.

Clearly, though, the firestream is going to be affected by the fact that the propellants do possess a high forward velocity at the time of release, and no allowance has been made for these effects. One obvious possibility is enhanced cooling due to mixing with ambient air. At speeds above Mach 1, shock effects may result in the existence of transient pressures and temperatures higher than those assumed here, but it is questionable whether these would last long enough at any one point to affect the vaporization. Bulk heating of the gases cannot be an important effect; if the entire kinetic energy of the released propellant went to heat the combustion gases, with none going to the ambient air, a velocity of about 1900 feet/second would be required to increase the temperature 10° .

Velocities this high are not reached below about 50,000 feet, and there is no pretense that the model has any meaning at much higher altitudes.

APPENDIX E
RECONDENSATION

Appendix E

Recondensation

As the temperature of the combustion gases following an abort declines, the equilibrium vapor pressure p_s will fall until it is less than the partial pressure p of vapor in the cloud. At this point, recondensation can begin, in principle. In practice, a sizable value of the supersaturation ratio $S \equiv p_s/p$ is required before liquid or solid particles can spontaneously precipitate out of the gas phase, especially if the vapor is very dilute as will be the case here. If inactive particles of other materials are present vapor can condense upon those also. The amount of supersaturation required for this process depends upon the degree of interaction between the vapor and the substrate material, but usually it is less than the supersaturation required for homogeneous condensation. If such condensation nuclei are available, heterogeneous condensation will therefore normally begin before homogeneous nucleation can occur, and if it proceeds rapidly enough it will deplete the cloud of vapor so that homogeneous nucleation never does occur. In what follows, we consider the heterogeneous process first.

Heterogeneous Condensation

For the most part, we will be considering particles of a few microns or less in diameter here. For these, the rate of recondensation (or evaporation) is governed primarily by the rate at which molecules can arrive at (or leave) the surface, rather than by diffusion through a boundary layer (Appendix A). In this case, the net rate of condensation on the surface of a particle is given by rewriting equation (A-2) with p_b set equal to p :

$$w(\text{g/sec}) = 44.34\alpha(p - p_s)(M/T)^{\frac{1}{2}}s \quad (\text{E-1})$$

where s is the particle surface area and α is the "sticking coefficient." If we now let w represent the rate at which the vapor concentration (by weight) is reduced due to condensation, n the number of particles per cm^3 , and $\bar{s}$ the arithmetic mean surface area per particle and note that $dp/dt = -w \cdot (RT/M)$, we may write

$$\frac{dp}{dt} = -44.34(p - p_s)R(T/M)^{1/2}\alpha n\bar{s}, \quad (\text{E-2})$$

where R is the gas constant. If p_s were to be held constant, then $d(p - p_s)/dt = dp/dt$ and we could write

$$\frac{d\ln(p - p_s)}{dt} = -44.34R(T/M)^{1/2}\alpha n\bar{s} \quad (\text{E-3})$$

Thus, the difference between p and p_s tends to decay out exponentially, provided other factors remain constant. Furthermore, one may replace $p - p_s$ by p in this relation if one has $p \gg p_s$, which must be the case before homogeneous condensation can begin to compete.

According to equation (E-1), the rate of condensation on each particle will be proportional to the particle's surface area throughout the process; hence, the final amount will be similarly proportional to the surface area, assuming the volume of the deposited PuO_2 is small compared with the total particle volume. The activity for a given particle will then be equal to $C_0 s / n\bar{s}$, where C_0 is the initial vapor concentration in activity units, provided all particles have similar values of α . The actual value of α is unknown; all numerical calculations in this work were performed with $\alpha = 1$.

The number and size distribution of particles is not known in the liquid-propellant abort case. Primarily as an example, we can perform a simple calculation using a representative size distribution quoted for soot in smoky air¹⁵, for which a count median diameter of .075 microns and a mass-median diameter of 0.5 microns were reported. If a log-normal size distribution is assumed, this corresponds to a value of $\ln \sigma_g$ equal to 0.795. From the properties of the log-normal distribution²², one then obtains a mean surface area of $6.3 \times 10^{-10} \text{ cm}^2$ and a mean mass of $7.6 \times 10^{-15} \text{ g}$, assuming a density of two for the particle material. If there are 10^{-8} g particles/ cm^3 in the fireball, one then obtains $n \approx 10^{-8} / 7.6 \times 10^{-15} = 1.3 \times 10^6$ particles/ cm^3 , $n\bar{s} = 8.2 \times 10^{-4}$. This density of particles corresponds to the conversion of less than 0.1% of the total carbon content into soot for the Saturn V propellant combination. When this result and other numerical values are inserted into equation (E-3), we obtain

$$\frac{d\ln(p - p_s)}{dt} \approx 8.2\alpha, \quad (\text{E-4})$$

for $T = 2000^\circ\text{K}$. Unless $\alpha \ll 1$ it is seen that this density of particles can scavenge the vapor rather rapidly.

For $^{238}\text{PuO}_2$ vapor, 12 Ci/gram, the activity concentration at 2000°K is 0.0197p curies/ cm^3 . With the particle size distribution given above, the mean activity per particle is $1.5 \times 10^{-8} \text{ p curies/particle}$. In the main text, it was shown that the vaporization of the entire SNAP-27 fuel load and uniform mixture of the vapor through a Saturn V fireball would yield $p \approx 8 \times 10^{-8} \text{ atm}$; hence, the mean particle activity would be about $1.2 \times 10^{-15} \text{ curies}$ or $1.2 \times 10^{-3} \text{ picocuries (pCi)}$. The "activity median" value would be somewhat larger, about $4.2 \times 10^{-3} \text{ pCi}$. That is, half of the total activity would

be in the form of particles more active than this and half would be in particles less active. The individual particle activities would then be distributed with $\ln \sigma_g = 1.59$ (twice the value for the diameters), and it is this distribution that was plotted in Fig. 15.

Actually, the value of p will vary through the vapor cloud and this will further broaden the activity distribution. In any case, the values of the parameters assumed here are essentially guesses. However, they are believed to be reasonable guesses and serve to illustrate the fact that the total activity will probably be distributed among a large number of particles with low individual activities. If the number density of particles were much lower than that assumed here (which would be required to obtain much higher individual activities), the recondensation would probably be largely by homogeneous nucleation.

Solid propellant fires differ from the liquid propellant case in that one of the major combustion products, Al_2O_3 , is a solid and condensation nuclei will be present in very large supply. Data on sizes are limited, but one set of measurements²⁰ on Al_2O_3 particles from a UTP-3001 propellant test* yielded mostly spherical particles with a roughly log-normal size distribution having a MMD in the range 0.21-0.24 microns, $\ln \sigma_g = 1.04$. From the properties of the log normal distribution²², one then obtains $\bar{s} = 2.2 \times 10^{-11} \text{ cm}^2$ and the mean mass is $1.9 \times 10^{-16} \text{ g}$. Since each gram of propellant produces about 0.3 g of Al_2O_3 and about 5700 cm^3 of combustion gas (at 2000°K), we obtain $n \approx 2.8 \times 10^{11} \text{ particles/cm}^3$, $n\bar{s} \approx 6.0$, and $d \ln(p - p_g)/dt = 6 \times 10^4 \alpha$ from equation (E-3). The very large numbers of

* It should be noted that this test was not an open-atmosphere burn; the size distribution resulting from the latter has not been measured.

particles will result in a very low activity per particle in the initial size spectrum for any reasonable value of p ; however, considerable coagulation must be expected as will be discussed briefly later.

Homogeneous Condensation:

In liquid-propellant aborts, it is at least conceivable that there may be inadequate foreign nuclei for heterogeneous condensation to deplete the vapor cloud before homogeneous condensation begins. Nucleation theory gives¹⁵ the nucleation rate J , in nuclei $\text{cm}^{-3}\text{sec}^{-1}$, to be

$$J = \frac{\alpha/2N^{3/2}}{\sqrt{\pi}R^2} \left(\frac{p}{T}\right)^2 \frac{\gamma M}{\rho} \exp \left[- \frac{16\pi M^2 \gamma^3}{3\rho^2 kT(RT \ln S)^2} \right] \quad (\text{E-5})$$

where $S = p/p_s$, α is the accommodation coefficient of the incipient droplet surface (taken here to be unity), N is Avogadro's number, γ is the surface tension, and k is the Boltzmann constant, and other quantities have been defined previously. Values of S and the corresponding values of T required to give $J = 1$ and $J = 10^6$ are listed in Table E-1 assuming $^{238}\text{PuO}_2$ vapor with $P = 8 \times 10^{-9}$, 8×10^{-8} , and 8×10^{-6} atm. Clearly, a substantial supersaturation is required in order to obtain significant nucleation rates, especially at the lower values of P .

Table E-1

$^{238}\text{PuO}_2$ Nucleation Rates

p	$J = 1 \text{ cm}^{-3}\text{sec}^{-1}$		$J = 10^6 \text{ cm}^{-3}\text{sec}^{-1}$	
	$T(^{\circ}\text{K})$	S	T	S
8×10^{-6}	2180	6.7	2142	11.6
8×10^{-8}	1851	16.1	1789	57
8×10^{-9}	1714	30	1625	257

Nucleation theory also gives an estimate for the initial radius of the incipient nucleus, which is

$$r_o = \frac{2\gamma M}{RT\rho \ln S} = 2.4 \times 10^{-8} \frac{\gamma M}{\rho T \ln S} \quad (E-6)$$

For the values of $\underline{S}$ in Table E-1, r_o is always very small, $< 10^{-7}$ cm. Subsequently, the particle is assumed to grow as in heterogeneous condensation, with the rate of mass addition per unit area given by equation (E-1).

Dividing by $\rho (=11.46)$ then gives the rate of size increase as

$$\frac{dr}{dt} = 44.34(p/\rho)(M/T)^{1/2} = 3.87p(M/T)^{1/2}, \quad (E-7)$$

where we have again assumed $\alpha = 1$.

Using equation (E-5) and the fireball time-temperature history for the Saturn V (Fig. 1), the nucleation rate was calculated as a function of time, for the values of $\underline{p}$ in table E-1. The initial particle size was assumed to be given by equation (E-6) and subsequent growth was governed by equation (E-7). The processes of particle growth and additional nucleation of new particles were followed until all the vapor initially assumed to be present had been condensed. The particle size distribution present at this time was converted into a distribution by activity and plotted in Fig. 17. Some mathematical approximations were made, but their effects are less than those due to more basic uncertainties such as the actual time-temperature history of the cooling fireball.

Coagulation:

If particle densities are high enough in a particulate cloud, coagulation must be expected. This will alter both the physical size distribution and the activity spectrum of a radioactive aerosol. The problems will only

be touched upon here, and some of the relations used will no doubt be applied outside their range of highest reliability. Nonetheless, it is possible to draw certain qualitative conclusions as to the maximum extent that coagulation can affect the nature of the aerosol.

First, we consider particles of roughly the same size, with $\underline{n}$ particles/cm³. In simplest approximation, coagulation reduces the number concentration at a rate

$$\frac{dn}{dt} = -Kn^2, \quad (E-8)$$

where $\underline{K}$ is the coagulation constant. Integrating then gives

$$\frac{1}{n} - \frac{1}{n_0} = Kt \quad (E-9)$$

where n_0 is the initial value of $\underline{n}$. If n_0 is large enough ($\gg 1/Kt$) we may then write

$$n \approx 1/Kt. \quad (E-10)$$

For particles of the same size, coagulation theory gives¹⁵

$$K = \frac{4RT}{3\eta N} \left(1 + \frac{A\lambda}{r}\right), \quad (E-10)$$

where λ is the molecular mean free path, r is the particle radius, and A is a constant with a value of about 0.9^{15} , and it has been assumed that the particles are spheres which coagulate if and only if they come into direct contact due to their Brownian motion. Inserting numerical values¹⁹ (for air) gives

$$\begin{aligned} K &= 2.95 \times 10^{-10} \left(1 + \frac{5.8 \times 10^{-6}}{r}\right) \text{ at } 288^\circ\text{K}, \\ K &= 6.21 \times 10^{-10} \left(1 + \frac{4.0 \times 10^{-5}}{r}\right) \text{ at } 2000^\circ\text{K}. \end{aligned} \quad (E-11)$$

In general, experimental data at ordinary temperatures are in good agreement with the first of these expressions¹⁵.

It appears, then, that $\underline{K}$ should be $\lesssim 10^{-9}$ cm³/sec for particles with $d \geq 1$ micron even at 2000°K, and $\leq 10^{-9}$ cm³/sec for particles down to 0.1 micron diameter once the gases have largely cooled. If we simply take $\underline{K}$ to be constant at 10^{-9} cm³, we find that coagulation will reduce the particle density to the order of $10^9/t$ particles/cm³ after $\underline{t}$ seconds, no matter how large the number density is initially.

The time over which coagulation can act is limited by the dispersal of the cloud due to mixing with ambient air. That process depends strongly upon meteorological conditions, but even a large fireball cloud is expected to be considerably diluted after 10^3 seconds, while observation of burn tests shows that the gases in the smoke column from a burning piece of solid propellant will be diluted substantially in less than ten seconds. Once this dilution process begins to reduce $\underline{n}$ at a rate comparable to coagulation, further coagulation will be greatly reduced. Atmospheric dilution does not affect the particle size and activity spectra, while the coagulation process does.

The above crude arguments indicate that coagulation will not reduce $\underline{n}$ below $10^6 - 10^8$ particles/cm³. If there are $\underline{q}$ grams of particles per cm³, and the density of the particle material is $\underline{\rho}$, the mean particle mass will be q/n , corresponding to a diameter of $(6q/\pi\rho n)^{1/3}$. If we assume $\rho \approx 2$ (as in carbon) and $n = 10^6$ for a liquid-propellant abort, the corresponding particle diameters will be $\sim .01q^{1/3}$ cm. Earlier calculations assumed $q = 10^{-8}$ g/cm³, which would give $d \approx 0.21$ microns. Of course, $\underline{q}$ could well be greater, but even $q = 10^{-5}$ g/cm³ would only give $d \approx 2$ microns. A value of $\underline{q}$ this high

would require conversion of the order of half the available carbon into soot, and combustion is unlikely to be that inefficient in any fire hot enough to cause much vaporization.

For solid-propellant fires, the aluminum loading ensures higher values of q , but it will still be $< 10^{-4}$ g/cm³. With $n \approx 10^8$, this value gives $d \approx 0.8$ micron.

The mean activity per particle will be $\approx .0197$ p/n curies, as noted before. For the above values of n , and any value of p that is at all reasonable, the mean activity will still be less than 1 pCi.

Assuming a monodisperse aerosol as was done here is a gross oversimplification. The actual situation will be much more complicated, with interactions between particles having a wide distribution of sizes, and furthermore, with a size distribution that is continually changing. There is, however, one feature of the process which may be noted by simple arguments.

We divide the size spectrum into a series of discrete ranges having radii $r_1, r_2, \dots, r_i, \dots, r_j, \dots$ and number densities $n_1, n_2, \dots$ etc. The rate at which particles of size r_i coagulate with particles of size r_j is given by²³

$$-\frac{dn(r_i, r_j)}{dt} = \frac{2}{3} \frac{RT}{N\eta} \frac{(r_i + r_j)^2}{r_i r_j} \left(1 + \frac{A\lambda}{r_{ij}}\right) n_i n_j, \quad (E-12)$$

where r_{ij} is an effective or "average" radius which is equal to $r_i r_j (r_i + r_j) / (r_i^2 + r_j^2)$; it is closer to the smaller of the two than the larger, especially if the size difference is large. If q_j is the mass/cm³ of particles in the j th size range, we will have $n_j = 4q_j / 3\pi r_j^3$, and hence

$$-\frac{dn(r_i, r_j)}{dt} = \frac{8RT}{9\pi\eta N} \frac{(r_i + r_j)^2}{r_i r_j} \left(1 + \frac{A\lambda}{r_{ij}}\right) n_i q_j \quad (E-13)$$

Depending upon the relative magnitudes of r_i and r_j , the rate at which particles of size r_i are removed by coagulation with particles of size r_j will show an inverse dependence upon r_j that is at least as strong as r_j^{-2} for a given value of q_j .

Now it is true by definition of the mass median diameter (MMD) that the sum of the q_j 's for $r_j > \text{MMD}/2$ must be equal to the sum of the q_j 's for $r_j < \text{MMD}/2$. However, the r_j^{-2} dependence weights in favor of the smaller particles. Thus most of the particles of size r_i will combine with particles smaller than the MMD rather than larger. On a mass-weighted basis, the process tends to reduce the number of particles by combining the smaller ones with each other more than it does by adding them onto the largest ones. To put it another way, those particles which are initially heaviest will tend to gain less mass, proportionately, than the smaller ones. If the initial size spread is very wide, it will tend to decrease, rather than increase, on a logarithmic scale⁽²³⁾. Though a substantial size spread must still be expected, the coagulation process is unlikely to yield a distribution so heavily skewed that most of the mass or radioactivity is concentrated in a small number of particles which are very much larger than the mean values given previously.

While the preceding arguments are anything but rigorous, they do suggest that coagulation is unlikely to result in a large fraction of the total activity being in particles large enough to be non-respirable or large enough so that they will not undergo long-range atmospheric transport. Likewise, it is unlikely to concentrate much of the activity into particles having individual activities $\gg 1$ pCi. The first of these conclusions, at least, simply corresponds to the general observation that most of the smoke

from a fire usually drifts off with the breeze to pollute the general environment, rather than coagulating into particles large enough to fall out quickly.

APPENDIX F
LIST OF SYMBOLS

Appendix F

List of Symbols

a	Radius (used only where r would cause confusion), (cm)
C	Concentration of vapor (g/cm^3)
C_b	Vapor concentration in a boundary layer
C_s	Vapor concentration at the saturated vapor pressure
C_∞	Vapor concentration at an infinite distance
C_D	Drag coefficient
$c_p (c_v)$	Specific heat at constant pressure (volume), ($\text{cal g}^{-1} \text{deg}^{-1}$)
$C_p (C_v)$	Molar specific heats at constant pressure (volume), ($\text{cal mole}^{-1} \text{deg}^{-1}$)
d	Diameter (cm)
D	Diffusion coefficient ($\text{cm}^2 \text{sec}^{-1}$)
f	Fraction vaporized
g	Acceleration of gravity, 980 cm sec^{-2}
h	Altitude (of a rocket booster vehicle)
H	Height above the surface of a piece of burning solid propellant (cm)
J	Nucleation rate (nuclei $\text{cm}^{-3} \text{sec}^{-1}$); diffusion current ($\text{g cm}^{-2} \text{sec}^{-1}$)
k	Boltzmann constant ($1.3804 \times 10^{-16} \text{ erg deg}^{-1}$)
K	Thermal conductivity ($\text{cal deg}^{-1} \text{cm}^{-1} \text{sec}^{-1}$); coagulation constant ($\text{cm}^3 \text{sec}^{-1}$)
$\ln$	Natural logarithm
$\log_{10}$	Base - 10 logarithm
L	"Characteristic length" indicating size of a solid propellant fragment

m	Mass (g)
M	Molecular weight (g mole ⁻¹)
n	Number density of an aerosol (particles cm ⁻³)
n _o	Value of <u>n</u> at time of formation of an aerosol
N	Avogadro's number, 6.0247 x 10 ²³ mole ⁻¹
p	Partial pressure of a vapor (atm)
p _s , p _b , p _∞	analogous to C _s , C _b , C _∞
P	Ambient atmospheric pressure (atm)
q	Weight density of an aerosol (grams of particles/cm ³)
r	Radius; radial distance (cm)
r _o	Initial radius of an incipient condensation nucleus (cm)
R	Gas constant, 8.3166 x 10 ⁷ erg deg ⁻¹ mole ⁻¹ = 82.057 cc atm deg ⁻¹ mole ⁻¹ = 1.9877 cal deg ⁻¹ mole ⁻¹
Re _d	Reynolds number for a sphere, = vd/ν (dimensionless)
s	Surface area of a particle (cm ²)
$\bar{s}$	Mean surface area of particles in an aerosol
S	Supersaturation ratio, = p/p _s
Sc	Schmidt number, = ν/D (dimensionless)
t	Time (sec)
t _r	Time interval between initiation of an abort and release of radioisotopic fuel
T	Temperature (°K)
v	Velocity of a particle with respect to the surrounding gases (cm sec ⁻¹)
v _b	Velocity of a booster vehicle
v _g	Velocity of combustion gases with respect to the general environment

v_p	Velocity of a particle with respect to the general environment
v_t	Terminal fall velocity
V	Volume (cm^3)
V_o	Nominal volume of a Saturn V fireball
w/A	Rate of weight loss (gain) per unit area ($\text{g cm}^{-2} \text{sec}^{-1}$)
W	Weight of propellant on board a booster vehicle
$\dot{W}$	Rate of spillage of propellants in an abort
α	Accommodation coefficient or "sticking coefficient"
γ	Surface tension (dyne/cm)
ϵ	Emissivity
η	Dynamic or "ordinary" viscosity ($\text{g cm}^{-1} \text{sec}^{-1}$)
λ	Molecular mean free path in a gas (cm)
ν	Kinematic viscosity, $= \eta/\rho$ ($\text{cm}^2 \text{sec}^{-1}$)
ρ	Density of a solid or liquid (g/cm^3)
ρ_g	Density of a gas (g/cm^3)
σ	Stefan - Boltzmann constant, $1.355 \times 10^{-12} \text{ cal deg}^{-4} \text{ cm}^{-2} \text{sec}^{-1}$
σ_g	Geometric standard deviation
ϕ	Heat flux, $\text{cal cm}^{-2} \text{sec}^{-1}$

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