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AN INFORMAL REPORT

Source Term Calculations for Pioneer Radioisotopic Thermoelectric Generators

MASTER

los alamos
scientific laboratory
of the University of California
LOS ALAMOS, NEW MEXICO 87544

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Source Term Calculations for Pioneer Radioisotopic Thermoelectric Generators

by

S. E. Bronisz

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and the Space Nuclear Systems Office.

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SOURCE TERM CALCULATIONS FOR
PIONEER RADIOISOTOPIC THERMOELECTRIC GENERATORS

by

S. E. Bronisz

ABSTRACT

Quantities of radioactive material that would be expected to be released as the result of the exposure of a Pioneer radioisotopic thermoelectric generator to certain accident environments have been calculated. The conventions used in these calculations are described in detail, as are the reasons for choosing the conventions. The final source terms are believed to be safety conservative, but the actual degree of conservatism cannot be determined.

1.0 INTRODUCTION

The evaluation of risks involved in using nuclear systems for space missions requires that the possible release of radioactive materials, the source term, to the biosphere be defined for each possible accident environment. The purpose of this report is to present the methods by which such source terms were calculated for accidents involving the nuclear power systems to be used in generating electrical power on the Pioneer spacecraft.

The Pioneer spacecraft is an interplanetary probe designed to fly past Jupiter and to examine interplanetary space. Power for the various experiments aboard the spacecraft will be provided by four radioisotopic thermoelectric generators (RTG's), each containing about 20,200 Ci of ^{238}Pu as plutonium dioxide combined with 17.5 vol% molybdenum in the form of cermet disks (PMC) contained within a metallic capsule. The capsule, in turn, is contained within a graphite heat shield. The fuel/capsule/heat shield assembly, i. e. the heat source,

is surrounded by thermoelectric elements and is contained with them in a gas-tight magnesium alloy housing. ⁽¹⁾

The heat source capsule is a cylinder with hemispherical ends made in four layers; an inner liner, a liner, a strength member, and an outer clad. The inner liner is Mo-46 wt% Re, 0.005 in. thick, and impedes interaction between the PuO_2 in the fuel and the tantalum components in the capsule. It is formed from three pieces, two hemispherical ends and a cylindrical body, by spot welding and has no structural effect on the capsule. The liner is Ta-20 wt% W, 0.02 in. thick, with a pressure relief device in one end. It is assembled by welding two pieces, a hemispherical cap and a cylindrical body with a hemispherical end, and is gas tight, except for the PRD which allows escape of the helium evolved from the decay of ^{238}Pu . The liner serves as a contamination barrier in the assembly operation. The third layer, the

strength member, is a 0.09-in. thick Ta-8 wt % W-2 wt % Hf alloy (T-111) shell designed to provide the primary mechanical protection. It, too, is assembled from two pieces by welding. The cap has two small holes to allow helium to escape. The outer layer, the clad, is made, again in two pieces, from 0.02 in. -thick Pt-20 wt % Rh. It has one small hole in the body end for helium release.

The responses of the heat source capsule and the fuel to thermal and mechanical environments representative of accident conditions were determined experimentally through use of capsules loaded with a fuel simulant. The results of the experiments were then used to predict the expected behavior of actual heat source capsules in possible accident environments.

No attempt is made here to integrate the source term calculations with the probability of occurrence of the environment responsible for the source term. This has been done in the Pioneer F Safety Evaluation Report. ⁽²⁾

3.0 ACCIDENT MODELS

Four general accident cases were considered: launch pad abort, air destruct, high velocity impact, and reentry impact. Each accident case includes a series of environments, present at various times during the accident, which are described below.

2.1 Launch Pad Abort

The basic assumption for this type of accident is that the liquid-fueled Atlas-Centaur launch vehicle explodes at the approximate time of launch resulting in exposure of the capsule to one or more of the following environments: blast overpressure, impact of fragments, fireball, impact on the ground at ~150 fps, and residual propellant fires. Except for the case of exposure to residual fires, the capsule temperature would be ~1500°F. The effects of the environments on a capsule and the total number of environments affecting the capsule depend on the stage of the accident at which the capsule is

separated from the RTG. The source terms presented below are based on the assumption that the capsule is exposed by the blast overpressure, but they are described in such a way that results of exposure at any later stage of the accident can be calculated.

2.2 Air Destruct Accident

This accident case would occur if the launch vehicle were to go off course sufficiently to warrant its destruction, as a protective measure, by the Range Safety Officer. Such an accident might expose the capsule to blast overpressure, fragment impact, fireball, ground impact at ~325 fps, and/or residual propellant fires. Again the assumption is that the blast-overpressure would expose the capsule. The capsule would be a temperature of ~1500°F, except when exposed to a residual fire.

2.3 High Velocity Impact

This accident would occur if a launch vehicle malfunction resulted in a 180° change in direction of the vehicle and a powered impact into the launch area without successful destruction action. As a result of such an accident, a heat source capsule might be exposed to impact, blast overpressure, fragment impact, fireball, and/or residual propellant fires. The impact velocity of the vehicle would be ~550 fps, but the capsule itself would have a lower impact velocity because of the mitigating effects of the generator, the spacecraft, and the rocket shrouds.

2.4 Reentry

In the event that the spacecraft should fail to reach escape velocity it would reenter the atmosphere. The heat of reentry would release the heat source from the generator and allow it to impact the earth. Thus, the capsule would be exposed to reentry heating (in the graphite heat shield), earth impact, and post impact oxidation. If the graphite heat shield were to fail, the unprotected capsule could fail in reentry and expose the bare fuel disks to the reentry and impact conditions. The impact velocity of an intact heat source would be ~300 fps

and that of released disks would be ~ 125 fps.

3.0 TEST RESULTS

The Pioneer heat source capsule was subjected to a series of tests designed to simulate environments thought to be representative of those attributed to the various possible accidents. The test series was designed by Teledyne Isotopes, the RTG contractor, and was carried out by personnel of Sandia Corporation. After being tested the capsules were examined at LASL to determine the response of the capsules and the fuel simulant to the tests. Details of the tests and the results are included in the test report documents published by Teledyne Isotopes. (3)

In addition to the capsule tests, a series of bare fuel impact tests was run to determine the correlation between the break-up characteristics of the fuel and its simulant. These tests were necessary because it was not practicable to test fueled capsules.

3.1 Capsule Response

The capsules used in the safety evaluation tests did not contain the 0.005-in. Mo-Re inner liner. The assumption has been made that the inner liner would not significantly affect the mechanical response of the capsule.

3.1.1 Overpressure Tests. Two capsules, each contained in a generator, were exposed to overpressures of 320 psig at 1.95 psig-sec and 1700 psig at 0.26 psig-sec, respectively. In both cases, the generator housing and the graphite heat shield assembly were removed by the blast, but the capsule was not breached. In the 320 psig test, the capsule appeared to be unaffected, while in the 1700 psig test the side of the capsule nearest the blast was deformed. The test results indicated that the capsule could withstand at least 1700 psig at 0.26 psig-sec without failing. This overpressure is thought to simulate the actual overpressure of the maximum realistic launch abort explosion. Since the capsule survived these tests, the conclusion is that it

would survive all credible overpressures and thus there are no overpressure source terms. Because the capsules were to be used in additional tests, the fuel response was not determined in the overpressure tests.

3.1.2 Fragment Impact Tests. Four capsules were subjected to impacts of four different fragments, a 38.5-lb electronic chassis at 409 fps, a 0.85-lb titanium sheet flat-on at 2100 fps, a 0.85-lb titanium sheet edge-on at 1700 fps, and a 1.28-lb titanium sheet edge-on at 495 fps. None of these fragments breached the capsule, but all except the flat-on sheet broke the outer cladding. Each of the fragments caused plastic deformation of the capsule and some break-up of the fuel simulant in the capsules.

For the source term analysis, it was assumed that no fragment impact would cause release of the fuel in the launch abort and air destruct accidents. It was further assumed, however, that, in the high velocity impact case, fragments having greater energies might be present and could damage the capsule sufficiently to cause release of fuel. Since no experimental failure was available to use as a basis for predicting the results of such an event, it was assumed that the impact of a high energy fragment would make as large a hole in the strength member as was made in the worst failure experienced in the granite impact test program described below.

3.1.3 Fireball. No tests to simulate exposure of the capsule to the fireball were made. Comparison of fireball conditions with those of the solid fire tests indicated that, because of the short duration of the fireball and the relatively large heat capacity of the capsule, no serious damage would be caused by the fireball.

3.1.4 Residual Propellant Fires. Four capsules were exposed to both liquid and solid propellant fires. The liquid fires caused no additional damage to any capsule and so it was concluded that exposure of a fueled capsule to a liquid propellant

fire would not result in capsule damage or fuel release.

In the solid fire tests, the capsule was damaged when not protected by its graphite heat shield, but not damaged when it was in the heat shield. The clad of a bare capsule placed between burning blocks of solid propellant melted and reacted with the strength member, but the capsule was not breached. Two bare capsules placed on a burning block of solid propellant, however, were breached, exposing fuel simulant, during a 4.75-min fire. In both capsules, the breaches were similar in size and location, being elliptical with major and minor axes of ~ 0.75 and ~ 0.5 in., respectively, and located in the hemispherical end of each capsule.

3.1.5 Impact. Test capsules were impacted on granite and hard soil at 300 to 348 fps at 2400°F and at various angles. The designated test velocity of 330 fps was chosen because it is about 10 percent higher than the terminal velocity of the capsule. Most of the capsules were exposed to a simulated reentry environment prior to impact.

The soil impact caused no capsule failure and only minor capsule deformation, so it was concluded that no capsule failures would occur in any accident environment involving soil impact.

Three out of seven of the capsules that were impacted on flat granite failed. The failure, in each case, consisted of a small crack about 0.06 in. $\times$ 0.75 in. extending through all three layers of the test capsule.

One capsule was impacted on chunks of granite, and failed through formation of a rough hole having an area of 2.5 cm^2 . The largest sphere that could pass through this hole would be 0.6 cm dia.

In the Transit program, ⁽⁴⁾ three capsules were impacted on concrete. None failed, so it was concluded that impacts on concrete would release no fuel.

On the basis of these results it was assumed that the fueled capsule would fail during a launch

abort only if it impacted the sharp edges of the steel launch structure. Also, failure was assumed in the air destruct case or in the high velocity impact case if the capsule impacted a hard surface.

Finally, failure was assumed if the capsule impacted a hard surface after reentry. In all cases, the extent of the failure was assumed to be equal to that of the largest failure seen, the impact onto rough granite. Such an assumption is conservative in all cases except that of the impact on rough, hard surfaces after reentry, but there seems to be no way of accurately scaling the magnitude of the failures to be expected in the other cases.

3.1.6 Post-impact Oxidation. In the Transit test program, impacted capsules still containing the fuel simulant were oxidized using both internal ⁽⁵⁾ and external ⁽⁴⁾ electrical heat sources. The results showed that after a few days the tantalum components oxidize, force the clad to split, and expose the fuel to the environment. This result is to be expected, even if the clad were not damaged, because the vent hole in the clad would permit atmospheric oxygen to contact the strength member.

3.2 Capsule Condition

All of the above statements on probable capsule response apply to capsules similar to those used in the test series. In the earlier capsule design, which included no molybdenum-rhenium inner liner, oxygen could migrate from the fuel into the tantalum liner and strength member, rendering them brittle. The addition of the molybdenum-rhenium inner liner apparently has eliminated this problem for the launch accident and prompt reentry accident. ^(6, 7) In the case of the delayed reentry accident, however, it must be assumed that the capsule will be brittle and will behave differently from the test capsules. No data are available to indicate the extent of the difference, but it was assumed that after several months the capsule would have essentially no fuel containment capability under accident conditions.

3.3 Fuel Response

3.3.1 Correlations. It was not possible to determine, directly, the behavior of PMC fueled capsules in the accident environments. Instead, the capsules were filled with a ThO_2 -14 wt % Mo fuel simulant. The particle size distribution of the simulant was determined after various tests, thus providing some of the information necessary to calculate source terms. (8)

A correlation between the fines distributions of fuel and simulant was determined by impacting small samples of each at 325 fps and 454°C (850°F). The correlation factors are given in Table 3.1. It is assumed that these factors also apply at higher temperatures. For all accident environments the temperature would be at least 815°C (1500°F). Since the melting point of the ThO_2 in the SSCS is about 900°C (1620°F) higher than that of the PuO_2 in the PMC, it is expected that at any temperature the PuO_2 would tend to be less brittle than the ThO_2 and, thus, the factors are conservative.

For the purpose of simplifying the calculations of the source terms the correlation factor for all launch cases was assumed to be 1.0, so the particle size data determined from the SSCS in the capsules could be used directly. For the case of delayed orbital reentry the factor was assumed to be 1.5.

One further assumption, that the quantity of particles formed on impact is proportional to the square of the impact velocity, was made. This

TABLE 3.1
PMC/SSCS IMPACT FINES CORRELATION
FACTORS, $\leq 4 \mu\text{m}$

PMC Condition	Multiply SSCS Fines By:
Fresh ^a	0.34
Aged ^b	0.83
Aged and Reentered ^c	1.03
Aged, long term ^d	1.47

^a No heat treatment after fabrication.

^b 82 days at 815°C.

^c 42 days at 815°C, followed by a simulated orbital reentry heat pulse.

^d 75 days at 1370°C.

allows the size distribution observed after an impact at one velocity to be corrected to apply to another velocity by multiplying the observed distribution by the quotient of the velocities squared.

3.3.2 Test Response.

3.3.2.1 Vibration. Each RTG is subjected to a qualification vibration test prior to launch and also to the vibration of the launch itself. Such vibration causes minor fracturing of the edges of the fuel disks, because the fuel does not completely fill the capsule. The amount of break-up to be expected was determined by comparing the particle size distributions of test capsules P-23 and P-24. Each capsule had been exposed to a reentry simulation and an impact at terminal velocity on soil. In addition, capsule P-23 had been subjected to a qualification level vibration test prior to impact, so that the differences between the fuel simulant responses could be used as a measure of the amount of break-up caused by the qualification level vibration test. The P-23 particle size data are given in Tables 3.2 and 3.3 and those of P-24 are given in Tables 3.4 and 3.5.

3.3.2.2 Overpressure and Fragment Impact. These two environments were not treated separately in the post-test examinations, but the fuel simulant from capsule P-13, which had been exposed to the 1700 psig overpressure and

TABLE 3.2
PARTICLE SIZE DISTRIBUTION,
PIONEER CAPSULE P-23

Impact velocity: 325 fps		Impact angle: 0.0°
		Impact temperature: 2400°F
Screen Size, Micron	Weight Fraction	Accumulated Weight Fraction
2000	0.99760	0.99760
841	.00016	.99776
420	.00017	.99792
177	.00016	.99808
125	.00008	.99816
74	.00016	.99831
44	.00025	.99856
30	.00022	.99878
20	.00027	.99905
10	.00040	.99945
-10	.00055	1.00000

TABLE 3.3

SUB-SIEVE SIZE PARTICLE DISTRIBUTION,
PIONEER CAPSULE P-23Impact velocity: 325 fps
Impact temperature: 2400°F

Maximum Size In Range, Microns	Number	Weight Fraction	Accumulated Weight Fraction
0.10	805	0.000000	0.000000
.20	439	.000000	.000000
.30	247	.000000	.000000
.40	159	.000000	.000001
.50	119	.000001	.000001
.60	92	.000001	.000002
.70	80	.000001	.000003
.80	57	.000001	.000004
.90	42	.000001	.000005
1.00	22	.000001	.000006
1.25	41	.000003	.000009
1.50	42	.000005	.000014
1.75	28	.000006	.000020
2.00	23	.000007	.000027
2.25	9	.000004	.000031
2.50	8	.000005	.000035
2.75	11	.000008	.000044
3.00	7	.000007	.000051
4.00	8	.000019	.000070
5.00	13	.000060	.000130
6.00	8	.000064	.000194
7.00	7	.000089	.000283
8.00	2	.000038	.000321
9.00	3	.000081	.000402
10.00	4	.000148	.000550

the 2100 fps flat sheet fragment impact, was subjected to a particle size analysis. The results are given in Tables 3.6 and 3.7. The size distribution observed was assumed to be characteristic of those for all fragment impacts.

TABLE 3.4

PARTICLE SIZE DISTRIBUTION,
PIONEER CAPSULE P-24Impact velocity: 325 fps Impact angle: 0.0°
Impact temperature: 2400°F

Screen Size Microns	Weight Fraction	Accumulated Weight Fraction
2000	0.99979	0.99979
841	.00008	.99987
420	.00003	.99990
177	.00001	.99992
125	.00001	.99993
74	.00001	.99994
44	.00001	.99995
30	.00001	.99996
20	.00001	.99997
10	.00002	.99999
-10	.00001	1.00000

TABLE 3.5

SUB-SIEVE SIZE PARTICLE DISTRIBUTION,
PIONEER CAPSULE P-24Impact velocity: 325 fps
Impact temperature: 2400°F

Maximum Size in Range, Microns	Number	Weight Fraction	Accumulated Weight Fraction
0.10	781	0.000000	0.000000
.20	528	.000000	.000000
.30	144	.000000	.000000
.40	78	.000000	.000000
.50	76	.000000	.000000
.60	71	.000000	.000000
.70	88	.000000	.000000
.80	51	.000000	.000000
.90	41	.000000	.000000
1.00	25	.000000	.000000
1.25	63	.000000	.000000
1.50	36	.000000	.000000
1.75	26	.000000	.000000
2.00	16	.000000	.000000
2.25	12	.000000	.000000
2.50	4	.000000	.000000
2.75	10	.000000	.000001
3.00	8	.000000	.000001
4.00	9	.000000	.000001
5.00	5	.000000	.000001
6.00	8	.000001	.000002
7.00	2	.000000	.000002
8.00	2	.000001	.000003
9.00	3	.000001	.000004
10.00	12	.000006	.000010

3.3.2.3 Impact. The extent of fuel

break-up in the Pioneer capsule is governed by the capsule attitude on impact, velocity, temperature, and the type of impact surface. In the accident cases assumed, two attitudes, two velocities, one

TABLE 3.6

PARTICLE SIZE DISTRIBUTION
PIONEER CAPSULE P-13Impact velocity: 2100 fps Impact angle: 0.0°
Impact temperature: 1850°F

Screen Size, Micron	Weight Fraction	Accumulated Weight Fraction
2000	.98421	.98421
841	.00937	.99358
420	.00214	.99572
177	.00144	.99716
125	.00038	.99754
74	.00053	.99807
44	.00058	.99865
30	.00030	.99895
20	.00030	.99925
10	.00034	.99960
-10	.00040	1.00000

TABLE 3.7
SUB-SIEVE SIZE PARTICLE DISTRIBUTION,
PIONEER CAPSULE P-13

Impact velocity: 2100 fps
Impact temperature: 1850°F

Maximum Size in Range, Microns	Number	Weight Fraction	Accumulated Weight Fraction
0.10	494	0.000000	0.000000
.20	267	.000000	.000000
.30	214	.000000	.000000
.40	115	.000000	.000000
.50	107	.000000	.000000
.60	117	.000000	.000000
.70	107	.000000	.000001
.80	79	.000000	.000001
.90	47	.000000	.000001
1.00	50	.000000	.000002
1.25	95	.000002	.000003
1.50	71	.000002	.000005
1.75	63	.000003	.000008
2.00	54	.000004	.000012
2.25	37	.000004	.000015
2.50	28	.000004	.000019
2.75	26	.000005	.000024
3.00	28	.000006	.000030
4.00	58	.000031	.000062
5.00	47	.000050	.000111
6.00	29	.000053	.000164
7.00	7	.000020	.000184
8.00	12	.000052	.000236
9.00	6	.000037	.000273
10.00	15	.000127	.000400

temperature, and two impact surfaces were considered.

The impact test program identified the end-on attitude as being the most damaging to the fuel, while the various drop tests identified the characteristic free fall impact attitude as being nearly side on. For the purpose of calculating source terms, the particle size data from capsule P-10, impacted end on, was used for the launch abort and high velocity impact accidents. These data are given in Tables 3.8 and 3.9.

For the air destruct and reentry accident cases, the heat source was assumed to be free to fall in its characteristic mode of flat spin. No tests were run in the Pioneer program to identify the fuel response to an impact on a hard surface at this attitude, so the particle size data for a Transit program capsule, T-35, impacted on granite with the capsule axis 22.5° from the plane of the

TABLE 3.8
PARTICLE SIZE DISTRIBUTION
PIONEER CAPSULE P-10

Impact velocity: 325 fps Impact angle: 0.0°
Impact temperature: 2400°F

Screen Size, Microns	Weight Fraction	Accumulated Weight Fraction
2000	0.90461	0.90461
841	.06110	.96571
420	.01399	.97970
177	.00622	.98592
125	.00148	.98740
74	.00264	.99004
44	.00202	.99206
30	.00219	.99425
20	.00138	.99563
10	.00168	.99731
-10	.00269	1.00000

impact surface were used. These data are given in Tables 3.10 and 3.11. The Transit and Pioneer capsules differ only in length. For impact on a relatively soft surface, the particle size data from capsule P-24, Tables 3.4 and 3.5, were used.

TABLE 3.9
SUB-SIEVE SIZE PARTICLE DISTRIBUTION,
PIONEER CAPSULE P-10

Impact velocity: 325 fps
Impact temperature: 2400°F

Maximum Size in Range, Microns	Number	Weight Fraction	Accumulated Weight Fraction
0.10	525	0.000000	0.000000
.20	434	.000001	.000001
.30	263	.000002	.000003
.40	257	.000005	.000008
.50	189	.000006	.000014
.60	131	.000008	.000022
.70	111	.000010	.000032
.80	81	.000011	.000044
.90	50	.000010	.000054
1.00	41	.000011	.000065
1.25	63	.000034	.000098
1.50	36	.000033	.000132
1.75	42	.000062	.000193
2.00	26	.000057	.000250
2.25	13	.000041	.000291
2.50	14	.000060	.000351
2.75	14	.000080	.000430
3.00	13	.000096	.000526
4.00	18	.000315	.000842
5.00	12	.000411	.001252
6.00	7	.000414	.001666
7.00	5	.000470	.002136
8.00	2	.000280	.002416
9.00	0	.000000	.002416
10.00	1	.000274	.002690

TABLE 3.10
PARTICLE SIZE DISTRIBUTION,
TRANSIT CAPSULE T-35

Impact velocity: 325 fps Impact angle: 22.5°
Impact temperature: 2400°F

Screen Size Microns	Weight Fraction	Accumulated Weight Fraction
2000	0.87804	0.87804
841	.08021	.95825
420	.01889	.97713
177	.00859	.98572
125	.00219	.98791
74	.00320	.99112
44	.00231	.99343
30	.00163	.99506
20	.00125	.99631
10	.00190	.99821
- 10	.00179	1.00000

Two velocities were considered, 150 fps for the launch abort accident and 325 fps for the other accidents. All impact tests were run at 325 fps, allowing the particle size distributions observed to be used directly for all accidents except the

TABLE 3.11

SUB-SIEVE SIZE PARTICLE DISTRIBUTION,
TRANSIT CAPSULE T-35

Impact velocity: 325 fps
Impact temperature: 2400°F

Maximum Size in Range, Microns	Weight Number	Weight Fraction	Accumulated Weight Fraction
0.10	421	0.000000	0.000000
.20	355	.000000	.000000
.30	237	.000000	.000000
.40	209	.000001	.000001
.50	140	.000001	.000002
.60	109	.000001	.000003
.70	91	.000001	.000004
.80	67	.000002	.000006
.90	40	.000001	.000007
1.00	45	.000002	.000009
1.25	59	.000005	.000014
1.50	38	.000006	.000020
1.75	20	.000005	.000024
2.00	25	.000009	.000033
2.25	17	.000009	.000042
2.50	17	.000012	.000053
2.75	11	.000010	.000064
3.00	20	.000024	.000087
4.00	38	.000107	.000194
5.00	30	.000165	.000360
6.00	19	.000181	.000540
7.00	25	.000378	.000918
8.00	17	.000383	.001301
9.00	7	.000225	.001526
10.00	6	.000264	.001790

launch abort accident. For the latter the characteristic impact velocity chosen was 150 fps and the characteristic impact attitude was end-on. The particle size distribution for these conditions was obtained by multiplying the distribution of P-10 by $(150/325)^2$ and is given in Tables 3.12 and 3.13.

The impact fines data were obtained at 1316°C (2400°F) for the capsule tests and at 454°C (850°F) for the bare fuel tests, and was assumed, as indicated above, that the correlations determined would apply to the whole range between these temperatures.

The fuel response to impact also depends on the hardness of the impact surface. For the purpose of calculating source terms, two impact surfaces, one hard and the other soft, were considered. Impact of the capsule on the hard surface was assumed to produce a particle size distribution like that of sample P-10 (Tables 3.8 and 3.9) or T-35 (Tables 3.10 and 3.11), whereas impact on a soft surface was assumed to produce a particle size distribution like that of sample P-24 (Tables 3.4 and 3.5).

In the event that the PMC fuel should escape from the capsule prior to ground impact, the released fuel would impact at approximately 125 fps, the free fall velocity of a PMC disk. In that case, the particle size distribution would be as shown in Table 3.14.

TABLE 3.12

CALCULATED PARTICLE SIZE DISTRIBUTION
FOR AN END-ON PIONEER CAPSULE
IMPACT ON GRANITE AT 150 fps

Size Microns	Weight Fraction	Accumulated Weight Fraction
2000	0.97968	0.97968
841	.01302	.99270
420	.00298	.99568
177	.00132	.99700
125	.00032	.99732
74	.00056	.99788
44	.00043	.99831
30	.00047	.99878
20	.00029	.99907
10	.00036	.99943
<10	.00057	1.00000

TABLE 3.13

CALCULATED SUB-SIEVE SIZE PARTICLE
DISTRIBUTION FOR AN END-ON PIONEER
CAPSULE IMPACT ON GRANITE AT 150 fps

Maximum Size in Range, Microns	Weight Number	Weight Fraction	Accumulated Weight Fraction
0.10	525	0.000000	0.000000
.20	434	.000000	.000000
.30	263	.000000	.000001
.40	257	.000001	.000002
.50	189	.000001	.000003
.60	131	.000002	.000005
.70	111	.000002	.000007
.80	81	.000002	.000009
.90	50	.000002	.000011
1.00	41	.000002	.000014
1.25	63	.000007	.000021
1.50	36	.000007	.000028
1.75	42	.000013	.000041
2.00	26	.000012	.000053
2.25	13	.000009	.000062
2.50	14	.000013	.000075
2.75	14	.000017	.000092
3.00	13	.000020	.000112
4.00	18	.000067	.000179
5.00	12	.000088	.000267
6.00	7	.000088	.000355
7.00	5	.000100	.000455
8.00	2	.000060	.000515
9.00	0	.000000	.000515
10.00	1	.000058	.000573

TABLE 3.14

CALCULATED PARTICLE SIZE DISTRIBUTION
FOR BARE PMC IMPACTED ON A HARD
SURFACE AT 125 fps

Size Microns	Weight Fraction	Accumulated Weight Fraction
2000	0.89904	0.89904
841	.03782	.93686
420	.02127	.95813
177	.01386	.97199
125	.00242	.97441
74	.00340	.97781
44	.00470	.98251
30	.00745	.98996
20	.00406	.99402
10	.00345	.99747
<10	.00253	1.00000
≤ 4	.00494	

$$\text{Fuel Release} = (0.037 \times W. F. \leq 6000 \mu\text{m} + 0.063 W. F. \leq 10 \mu\text{m}) \times 20,200 \text{ Ci} \quad (4.1)$$

3.3.2.4 Post-Impact Oxidation.

Once the fuel within the capsule has been exposed to air, its molybdenum component will oxidize, freeing the PuO_2 particles. Some of these particles will

be broken into fines on impact, even though surrounded by molybdenum. The quantity of additional fines available was determined by taking the particles $> 10 \mu\text{m}$ from an impacted and sized PMC sample, oxidizing them in flowing air for 10 days at 620°C (the calculated post-impact temperature), and sizing them again. The results showed that 35 percent of the residual was still larger than the ceramic shard size, so the oxidation was assumed to be 65 percent complete. The weight fraction $\leq 4 \mu\text{m}$ in the residue was calculated to be 0.001 and the total fraction possible, on complete oxidation, was calculated to be 0.00155. This latter term was used for source term calculations.

4.0 FUEL RELEASE ASSUMPTIONS

4.1 Mechanical Events

No data exist for the quantity of PMC released through a hole in a capsule, so it was assumed that all the fuel behind a hole would be released when the hole is formed, with the provisos that no piece larger than the hole could pass through it, and the hole would be considered empty for subsequent events. The size distribution of the fuel in the capsule was assumed to be uniform throughout the capsule. It was also assumed that every hole formed would be as large as the largest hole observed in the test program, 2.5 cm^2 , and large enough to pass a 0.6-cm dia piece. A hole this size accounts for 0.037 of the Pioneer capsule's longitudinal cross-sectional area. In addition, it was assumed that 10 percent of the particles $\leq 4 \mu\text{m}$ present would pass out through the hole.

The total release for each event causing a hole is:

The biologically important particle size is $\sim 4 \mu\text{m}$ because particles this size and smaller can become entrained in the lower lung. In order to enter the lung, however, they must first be in the breathing zone. Not all particles released from

the capsule will be immediately suspended in the breathing zone. Boring⁽⁹⁾ has estimated that the fraction $\leq 4 \mu\text{m}$ immediately suspended would range from 10^{-2} for low velocity events in still air to 10^{-1} for high velocity events in turbulent air. Still air conditions would not be expected to be present in the case of accidents near the launch area, but they might exist in the reentry accident case. In all cases considered here, however, the prompt resuspension factor was assumed to be 10^{-1} . Thus the prompt release of respirable particles to the breathing zone would be given by:

$$\text{Respirable Source Term} = [0.1 \times 0.1 \times \text{W. F.} \leq 4 \mu\text{m}] 20,200 \text{ Ci} \quad (4.2)$$

4.2 Thermal Events

4.2.1 Fireball. The fireball itself cannot cause a fuel release, but, if fuel is released by a mechanical event, the fireball can then contribute to the source term by vaporizing some of the mechanically released fuel. The amount vaporized depends on the size of the fuel particles in the fireball and can be calculated using Williams' model.⁽¹⁰⁾ Specifically, this model was derived from PuO_2 spheres $\sim 100 \mu\text{m}$ dia, but it has been extended to other sizes by Boring.⁽¹¹⁾ The weight fractions vaporized for the various sizes are given in Table

Original Particle Diameter, Microns	Weight Fraction Vaporized
6000	0.0007
2000	.0007
841	.0041
420	.016
177	.092
125	.185
74	.528
44	.957
≤ 30	1.000

The quantity vaporized is the product of the PMC area exposed, the time, and the vaporization rate at the flame temperature. In vacuum, the vaporization rate at the 2500°K flame temperature is $1.35 \times 10^{-3} \text{ g/cm}^2\text{-sec}$ ⁽¹²⁾ and in air it is 0.02 of this⁽¹³⁾ or $0.27 \times 10^{-4} \text{ g/cm}^2\text{-sec}$. The specific activity of PMC is 11.7 Ci/g, so the vaporization rate is $0.32 \times 10^{-3} \text{ Ci/cm}^2\text{-sec}$.

In the case of a capsule in contact with a piece of burning solid propellant the area exposed is equal to the area of the hole in the capsule and the vapor released would be given by:

$$\text{Vapor Source Term} = 0.31 \times 10^{-3} \times \text{B. T.} \times A_{\text{Hole}} \text{ Ci} \quad (4.3)$$

4.1. Thus, the total quantity vaporized would be the sum of the products of the weight fraction released and the weight fraction vaporized for each size fraction, multiplied by the total activity in the capsule.

If no holes are present in the capsule, it is assumed that a 2.4 cm^2 hole forms after 45 sec and that the time available for vaporization through this hole is the burning time less 45 sec. Thus, the release would be given by:

$$\begin{aligned} \text{Vapor Source Term} &= 0.31 \times 10^{-3} \text{ Ci/cm}^2\text{-sec (B. T. - 45 sec)} \times 2.42 \text{ cm}^2 \\ &= 0.75 \times 10^{-3} \text{ (B. T. - 45 sec) Ci} \end{aligned} \quad (4.4)$$

4.2.2 Solid Propellant Residual Fire. The fuel release from the solid propellant fire is in the form of vapor from pieces of PMC that come in contact with burning pieces of the solid propellant.

5.0 FUEL CONDITION

The particle size distribution of the PMC fuel in a capsule depends on the mechanical events to which it has been exposed and is different for

each possible accident case. Obviously, every capsule would contain the vibration fines, but not all would be subjected to a fragment impact or a hard surface impact. The particular size distribution to be expected in each of the general accident cases is discussed below.

5.1 Launch Pad Abort

In this accident, the only fuel release as a result of the mechanical environment would be caused by impact on sharp steel. There are, therefore, only two size distributions to consider. One results from vibration (P-23, P-24) and 150 fps impact [the size distribution of P-10 multiplied by $(150/325)^2$], and the other results from vibration, fragment impact (P-13), and 150 fps capsule impact. These distributions are shown in Figure 5.1 and listed in Table 5.1. The particle size data were obtained only up to 2000 μm , so the fractions present of other, larger, sizes were determined by extrapolation, assuming an asymptotic approach to the spherical equivalent diameter of a PMC fuel disk.

5.2 High Velocity Accident

In this case, it was assumed that fuel would be released from the capsule by the impact and the fragment impact. The blast overpressure would cause all the fuel released from the capsule to be blown into the air where a portion of it would be vaporized by the fireball. Those fuel fragments larger than 420 μm would be in the fireball until the start of lift-off, 1.5 sec, while the smaller particles would be entrained in the fireball for its effective lifetime, 6 sec.

The particle size distribution of the fuel in the capsule would be that resulting either from vibration (P-23, P-24) and 325 fps impact (P-10) or from vibration, impact, and fragment impact (P-13). These size distributions are shown in Figure 5.2 and listed in Table 5.2.

5.3 Air Destruct Accident

In this accident it was assumed that fuel release would occur only on terminal impact onto a hard

Fig. 5.1. Fuel size distribution, launch pad abort accident.

surface. Again, there are two possible particle size distributions in the PMC fuel, one resulting from vibration (P-23, P-24) and 325 fps free fall impact (T-35) and the other from vibration, fragment impact (P-13), and 325 fps free fall impact. These size distributions are shown in Figure 5.3 and listed in Table 5.3.

5.4 Reentry

5.4.1 Capsule Intact. On terminal impact after reentry, fuel release will occur only on

TABLE 5.1

FUEL SIZE DISTRIBUTION--LAUNCH PAD ABORT
Weight Fraction $\leq D$

Size, D Microns	No Fragment Impact	Fragment Impact
6000	0.19 ^a	0.28 ^a
2000	.053 ^a	.084 ^a
841	.02254	.03832
420	.00944	.01585
177	.00632	.01059
125	.00485	.00768
74	.00446	.00691
44	.00375	.00567
30	.00308	.00442
20	.00240	.00344
10	.00185	.00259
<10	.00111	.00151
≤ 4	.000248	.000310

^aDetermined by extrapolation, see Fig. 5.1.

Fig. 5.2. Fuel size distributions, high velocity impact accident.

impact with a hard surface. There are, however, two fuel particle size distributions to consider, which depend on the age of the fuel. The particle size distribution will be determined by pre-launch vibration (P-23, P-24), launch vibration (P-23, P-24), and 325 fps free fall impact (T-35 or 1.5 x T-35). These size distributions are given in Figure 5.4 and listed in Table 5.4.

5.4.2 Capsule Failure. If the reentry conditions are such that the heat shield fails, the

TABLE 5.2
FUEL SIZE DISTRIBUTION--HIGH VELOCITY
ACCIDENT

Size D, Microns	Weight Fraction $\leq D$	
	No Fragment Impact	Fragment Impact
6000	0.62 ^a	0.73 ^a
2000	.23 ^a	.26 ^a
841	.09761	.11339
420	.03643	.04284
177	.02230	.02657
125	.01593	.01876
74	.01438	.01683
44	.01159	.01351
30	.00933	.01067
20	.00693	.00797
10	.00529	.00603
<10	.00323	.00363
≤ 4	.000911	.000973

^aDetermined by extrapolation, see Fig. 5.2.

Fig. 5.3. Fuel size distributions, air destruct accident.

capsule will also fail and release the fuel disks to reenter bare. During the reentry, some of the fuel will vaporize and the remainder will impact on the ground. The size distribution of the fuel impacting the ground is given in Table 3.14.

6.0 SOURCE TERMS

The possible source terms resulting from the various accidents considered here will now be described.

TABLE 5.3
FUEL SIZE DISTRIBUTION--AIR DESTRUCT
ACCIDENT

Size D, Microns	Weight Fraction $\leq D$	
	No Fragment Impact	Fragment Impact
6000	0.70 ^a	0.77 ^a
2000	.28 ^a	.33 ^a
841	.12418	.13996
420	.04389	.05030
177	.02486	.02913
125	.01612	.01895
74	.01386	.01631
44	.01051	.01243
30	.00796	.00930
20	.00612	.00716
10	.00461	.00535
<10	.00233	.00273
≤ 4	.000263	.000325

^aDetermined by extrapolation, see Fig. 5.3.

5.4. Fuel size distribution, intact capsule reentry.

6.1 Launch Pad Abort

The size distributions present in the capsule as a result of a launch pad accident are shown in Figure 5.1 and listed in Table 5.1. The total release from a capsule that fails on terminal impact would be, from Eq. (4.1):

No fragment impact:

$$[0.037 \times 0.19 + 0.063 \times 0.00111] 20,200 \text{ Ci} = 143 \text{ Ci}$$

Fragment impact:

$$[0.037 \times 0.28 + 0.63 \times 0.00151] 20,200 \text{ Ci} = 211 \text{ Ci}$$

TABLE 5.4

FUEL SIZE DISTRIBUTION-REENTRY IMPACT		
Weight Fraction ≤ D		
Size, D, Microns	Prompt Reentry	Delayed Reentry
6000	0.65 ^a	0.815 ^a
2000	.27 ^a	.37 ^a
841	.12640	.18742
420	.04603	.06694
177	.02686	.03832
125	.01797	.02513
77	.01564	.02170
44	.01214	.01660
30	.00935	.01265
20	.00730	.00978
10	.00553	.00738
<1	.00287	.00377
≤4	.000332	.000429

^a Determined by extrapolation, see Fig. 5.4.

The prompt dispersion of respirable sized particles, $\leq 4 \mu\text{m}$ dia, would be, using Eq. (4.2)

No fragment impact:

$$[0.1 \times 0.1 \times 0.000248] 20,200 \text{ Ci} = 0.05 \text{ Ci}$$

Fragment impact:

$$[0.1 \times 0.1 \times 0.00031] 10,200 \text{ Ci} = 0.06 \text{ Ci}$$

In the event that the capsule should land on a piece of solid propellant, the additional source term for the vapor formed would be determined from Eqs. (4.3) and (4.4). The vapor would be released near ground level.

6.2 High Velocity Accident

The size distributions present in the capsule for this accident are given in Fig. 5.2 and Table 5.2. The total activity released on failure would be:

- 1) impact failure with no fragment impact, 467 Ci;
- 2) impact failure with fragment impact, 550 Ci;
- 3) impact and fragment failures, 1100 Ci.

These quantities would be released to the fireball, and the resulting vapor source terms would be, using the releases above and the vaporization terms shown in Table 4.1, 14.6 Ci, 16.9 Ci, respectively. The vapor would rise with the fireball and be dispersed at the fireball's characteristic height.

An additional, near ground, vapor source term would be present if any PMC material released from the capsule but not consumed by the fireball should come into contact with burning solid propellant. The quantity vaporized would then be given by Eq. (4.3), and the area would be the exposed area of the PMC.

6.3 Air Destruct Accident

The size distribution of the fuel in a capsule at the time of failure is shown in Figure 5.3 and Table 5.3. The release on terminal impact with a hard surface would be 526 Ci if no fragment impact occurred, or 579 Ci if a fragment did impact the capsule. The prompt suspension of respirable fines would be 0.05 Ci and 0.07 Ci, respectively.

6.4 Reentry

6.4.1 Capsule Intact. The size distribution of the fuel would be one or the other of those given in Figure 5.4 and listed in Table 5.4, depending on the age of the fuel. The release, through the single hole postulated to form on impact with a hard surface, would be 489 Ci or 609 Ci. The prompt suspension should be 0.07 Ci or 0.09 Ci.

6.4.2 Capsule failure. Should the capsule fail on reentry, some portion of it would vaporize and the remainder would impact the ground. The resulting particle size distribution is given in Table 3.14. The total near ground release would be the product of the fraction remaining after reentry and the total capsule activity, 20,200 Ci. The fraction vaporized was calculated to be 0.16. (3) So, 3250 Ci would be released as vapor in the upper atmosphere and 16,950 Ci would impact the ground. The prompt suspension of respirable particles would amount to 0.56 Ci if the fuel were relatively fresh or 0.73 Ci if it were aged.

6.5 Post-Impact Oxidation

After oxidation of the capsule and the molybdenum component of the fuel, the fines present will be available for dispersion. The total weight fraction of fines present would be 0.00188 for prompt impacts and 0.00275 for delayed impacts, 38 Ci and 56 Ci, respectively. The time dependent distribution of the fines thus exposed has been calculated by Fuquay. (2, 14)

7.0 SUMMARY

The source terms calculated for each accident are summarized in Table 7.1. These terms were calculated using the assumptions described above. As previously indicated, the basic assumption is that the fueled capsules will respond in the same manner as did those used in the safety test series. If this assumption is correct, the source terms calculated here are realistic, but conservative, estimates of those to be expected for fueled capsules exposed to accident environments.

ACKNOWLEDGMENTS

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TABLE 7.1
SOURCE TERM SUMMARY FOR A PIONEER RTG EXPOSED TO
ACCIDENT ENVIRONMENTS LEADING TO CAPSULE FAILURE

Event, Release Conditions	Source Term, Ci		
	Vapor ^a	Respirable ^b	Non-Respirable ^c
1.0 Launch Pad Abort - Failure occurs if bare capsule impacts sharp steel.			
1.1 No fragment impact.	0	0.05	143.
1.2 With fragment impact.	0	0.05	211.
2.0 High Velocity Accident - Failure occurs for high velocity impact on steel and/ or by very high velocity fragment impact.			
2.1 No fragment impact.	14.6	0 ^d	452.
2.2 With fragment impact.	16.8	0 ^d	533.
2.3 With fragment impact failure.	33.8	0 ^d	1066.
3.0 Air Destruct Accident - Failure occurs if bare capsule impacts hard surface.			
3.1 No fragment impact.	0	0.05	526.
3.2 With fragment impact.	0	0.07	579.
4.0 Reentry-Intact - Failure occurs if capsule impacts hard surface.			
4.1 Prompt reentry.	0	0.07	489.
4.2 Delayed reentry	0	0.09	609.
5.0 Reentry-Failure - Capsule fails and releases fuel during reentry.			
5.1 Prompt reentry.	3250. ^e	0.56	16,949.
5.2 Delayed reentry.	3250. ^e	0.73	16,949.
6.0 Post-Impact Oxidation - Intact capsule fails and exposes fuel.			
6.1 Prompt reentry.	0	38 ^f	20,162
6.2 Delayed reentry.	0	56 ^f	20,144

^a The vapor source term refers to that material vaporized in a fireball. An additional vapor term due to direct contact with burning solid propellant may be present. Its magnitude would be given by Eq. (4.3) or (4.4).

^b The respirable source term is that quantity of fuel $\leq 4 \mu\text{m}$ diameter and suspended in the atmosphere promptly after a mechanical event.

^c The non-respirable source term is that quantity of fuel $> 4 \mu\text{m}$ that escapes the capsule and those particles $\leq 4 \mu\text{m}$ that are not promptly suspended in the atmosphere.

^d In this accident a fireball was assumed to be inevitable, so all the fines released would be vaporized.

^e This vapor would be released in the upper atmosphere during reentry. The quantity was estimated in Ref. 3.

^f This respirable source term is the total $\leq 4 \mu\text{m}$ fines available from an impacted and completely oxidized capsule. The release occurs over a period of weeks.