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CALCULATED FISSION PRODUCT RELEASE FROM PRTR RUPTURE

AUTHOR

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CALCULATED FISSION PRODUCT RELEASE FROM PRTR RUPTUREINTRODUCTION

Approximately 15 minutes after shutdown on August 21, 1962, evidence of a rupture was observed in the form of high radiation levels in the containment vessel and rupture monitor indications.

Discharge of a special MgO-PuO_2 test element revealed a cladding break of about 1.5 inches in length and about $1/4$ inch wide in the widest place, located 36 inches from the top of the fuel element as it would have been oriented in the reactor. A visual examination in Radiometallurgy revealed that approximately 9.25 inches of fuel material or about 0.55% of the entire fuel element had escaped.

SUMMARY

The quantity of fission products released from the rupture is presented in Table I.

TABLE ICONTAMINATION RELEASE SUMMARY

Grams Plutonium	1.81
Curies Plutonium*	3.11
Grams Fissionable Plutonium	1.6
Grams MgO	100
Curies Fission Products ($\beta + \alpha$):	
At shutdown	15,000
10-day cooling	390
30-day cooling	137
90-day cooling	44
180-day cooling	18

* A value of 1.72 curies/gram of plutonium was calculated for the isotopic percentages of the plutonium:

	<u>Percent</u>	<u>Half-life (years)</u>
Pu-239	86.83	24,360
Pu-240	11.61	6,760
Pu-241	1.44	13
Pu-242	0.085	$3.79(10)^5$

The quantities and radiation levels from the individual fission product isotopes are calculated and presented in Tables II, III, IV and V. Table II gives the curies of various isotopes released by the rupture for 10 days, 30 days, 90 days, and 180 days after the rupture occurred. Table III gives the radiation levels one foot from an unshielded point source containing the various isotopes.

Table IV gives the radiation levels one foot from a point source shielded by one inch of steel. Table V gives the surface dose rate for the 14-inch primary piping if each isotope were dissolved uniformly in the primary coolant. Fission products having less than a one-day half-life were ignored unless they had a long lived precursor.

TABLE II

FISSION PRODUCT ACTIVITY

Isotope	10-day Curies	30-day Curies	90-day Curies	180-day Curies	Half-life
Zn-72	0.00013	small	small	small	49 hr
Ga	0.000054	small	small	small	14.3 hr
As-76	small	small	small	small	26.6 hr
As-77	0.00875	small	small	small	38.8 hr
Se _m	0.000175	small	small	small	17.5 sec
Se-79	small	small	small	small	6.5(10) ⁴ yr
Br-82	0.0000137	small	small	small	35.9 hr
Kr-85	0.006	0.006	0.006	0.006	10.27 yr
Rb-86	0.00256	0.00112	0.000115	small	19.5 d
Sr-89	4.81	3.75	1.77	0.535	54 d
Sr-90	0.04	0.04	0.04	0.04	28 yr
Y	0.0585	0.0585	0.0585	0.0585	64.5 hr
Y-91	8.68	6.95	3.3	1.13	56 d
Zr-95	13.2	9.02	5.74	2.3	63 d
Nb _m	0.0707	0.127	0.0707	0.0283	90 hr
Nb	2.126	7.45	7.45	4.25	35 d
Mo-99	14.9	0.0744	small	small	67 hr
Te _m	2.45	0.014	small	small	6.04 hr
Ru-103	21.1	14.0	5.15	1.12	41 d
Rh _m	19.6	13.1	4.8	1.05	57 min
Rh-105	1.91	small	small	small	36.5 hr
Ru-106	1.90	1.86	1.62	1.39	1 yr
Rh	1.98	1.94	1.69	1.44	30 sec
Ag-110 _m	0.00053	0.000503	0.000398	0.000345	270 d
Ag-111	3.29	0.527	0.00197	small	7.6 d
Cd-115	0.00602	0.000012	small	small	53 hr
In _m	0.0677	small	small	small	4.5 hr
Sn-119 _m	small	small	small	small	275 d
Sn-121	0.00369	small	small	small	27.5 hr
Sb-122	small	small	small	small	2.75 d
Sn-123 _m	0.00497	0.00412	0.0035	0.00226	131 d
Sn-125 _m	0.67	0.156	0.00171	small	9.4 d
Sb	0.0122	0.0186	0.0186	0.0182	2.7 yr
Te _m	0.0000135	0.0000673	0.000135	0.00135	58 d
Sb-127	3.58	0.0935	small	small	93 hr
Te _m	0.203	0.406	0.284	0.142	90 d
Te	11.5	0.596	0.209	0.109	9.3 hr
Te-129 _m	3.43	2.06	0.583	0.0859	33 d
Te	0.931	0.638	0.186	0.0744	72 min
Te-131 _m	small	small	small	small	30 hr
Te	small	small	small	small	24.8 hr
I	25.5	4.56	0.0267	small	8.05 d
Xe _m	1.87	1.22	0.0562	0.000468	12 d
Te-132	18.4	0.23	small	small	77 hr
I	14.7	0.192	small	small	2.4 hr

TABLE II (Continued)

<u>Isotope</u>	<u>10-day Curies</u>	<u>30-day Curies</u>	<u>90-day Curies</u>	<u>180-day Curies</u>	<u>Half-life</u>
Xe-133 _m	0.437	0.00106	small	small	2.3 d
Xe	54.1	3.74	small	small	5.27 d
Cs-134	0.000102	0.0001	0.0001	0.00009	2.0 yr
Cs-136	0.741	0.232	0.0077	0.00008	13 d
Cs-137	0.114	0.114	0.114	0.114	26.6 yr
Ba _m	0.0946	0.0946	0.0946	0.0946	2.6 min
Ba-140	37.2	12.6	0.461	0.00341	12.8 d
La	50.7	17.1	0.645	0.00462	40.2 hr
Ce-141	22.1	14.8	4.33	0.585	32 d
Ce-143	1.48	small	small	small	33 hr
Pr	37.	13.6	0.622	0.0066	13.7 d
Ce-144	2.42	2.3	1.94	1.7	290 d
Pr	2.33	2.21	1.86	1.63	17.5 min
Nd-147	1.48	0.425	0.011	0.000044	11.3 d
Pm	0.221	0.315	0.382	0.302	2.6 yr
Pm-148	0.00187	0.00014	small	small	5.3 d
Pm-149	2.34	small	small	small	54 hr
Pm-151	0.0842	small	small	small	27.5 hr
Sm	0.00394	0.004	0.00394	0.0039	93 yr
Eu-152	small	small	small	small	13 yr
Eu-153	0.38	0.000314	small	small	47 hr
Eu-154	0.00005	0.00005	0.00005	0.00005	16 yr
Eu-155	0.0364	0.0356	0.0032	0.0301	1.78 yr
Eu-156	1.34	0.547	0.037	0.0006	15.4 d
Tb-160	small	small	small	small	73.5 d
Tb-161	0.0287	0.0387	0.00001	small	7.0 d
Total	392	137	43.6	18.2	

TABLE III

POINT SOURCE RADIATION LEVELS - NO SHIELDING

Isotope	30-day r/hr	90-day r/hr	180-day r/hr	Remarks
Zn-72	small	small	small	
Ga	0.0001	small	small	
As-76	small	small	small	
As-77	0	0	0	no γ
Se _m	small	small	small	
Se-79	0	0	0	no γ
Br-82	small	small	small	
Kr-85	small	small	small	
Rb-86	0.00087	0.0001	small	
Sr-89	0	0	0	no γ
Se-90	0	0	0	no γ
Y	small	small	small	
Y-91	small	small	small	
Zr-95	39.6	25.2	10.1	
Nb _m	0.179	0.0995	0.0398	
Nb	33.2	33.2	19	
Mo-99	0.109	small	small	
Te _m	0.0113	small	small	
Ru-103	39.9	14.6	3.19	
Rh _m	3.14	1.15	0.251	
Rh-105	small	small	small	
Ru-106	0	0	0	no γ
Rh	2.85	2.47	2.12	
Ag-110 _m	0.00785	0.0062	0.0054	
Ag-111	0.094	0.00035	small	
Cd-115	small	small	small	
In _m	small	small	small	
Sn-119 _m	small	small	small	
Sn-121	0	0	0	no γ
Sb-122	small	small	small	
Sn-123 _m	0	0	0	no γ
Sn	small	small	small	
Sn-125 _m	0.089	0.00097	small	
Sb	5.15	5.15	5.05	
Te _m	0.00004	0.00009	0.0009	
Sb-127	0.403	small	small	
Te _m	0.22	0.154	0.077	
Te	0	0	0	no γ
Te-129 _m	1.31	0.37	0.545	
Te	4.2	1.23	0.49	
Te-131 _m	small	small	small	
Te	small	small	small	
I	10.7	0.0625	small	
Xe _m	1.19	0.055	0.00046	
Te-132	0.321	small	small	
I	2.31	small	small	

TABLE III (Continued)

<u>Isotope</u>	<u>30-day r/hr</u>	<u>90-day r/hr</u>	<u>180-day r/hr</u>	<u>Remarks</u>
Xe-133 _m	0.0015	small	small	
Xe	1.82	small	small	
Cs-134	8.7	8	7.5	
Cs-136	1.26	0.042	0.00042	
Cs-137	0	0	0	no γ
Ba _m	0.375	0.375	0.375	
Ba-140	17.9	0.654	0.0048	
La	217	8.2	0.059	
Ce-141	8.6	2.52	0.034	
Ce-143	small	small	small	
Pr	0	0	0	no γ
Ce-144	0.593	0.5	0.438	
Pr	1.06	0.896	0.785	
Nd-147	0.603	0.0155	0.00006	
Pm	0	0	0	no γ
Pm-148	small	small	small	
Pm-149	small	small	small	
Pm-151	small	small	small	
Sm	0.00045	0.00045	0.00045	
Eu-152	small	small	small	
Sm-153	0.0003	small	small	
Eu-154	small	small	small	
Eu-155	0.15	0.135	0.127	
Eu-156	3.94	0.267	0.0043	
Tb-160	0	0	0	no γ
Tb-161	0.0163	0.00004	small	
Total	407	105	50	

TABLE IV

POINT SOURCE RADIATION LEVELS - ONE INCH STEEL SHIELDING

<u>Isotope</u>	<u>30-day r/hr</u>	<u>90-day r/hr</u>	<u>180-day r/hr</u>	<u>Remarks</u>
Zn-72	small	small	small	
Ga	small	small	small	
As-76	small	small	small	
As-77	0	0	0	no γ
Se _m	small	small	small	
Se-79	0	0	0	no γ
Br-82	small	small	small	
Kr-85	small	small	small	
Rb-86	0.00055	0.000055	small	
Sr-89	0	0	0	no γ
Sr-90	0	0	0	no γ
Y	0.0013	0.0013	0.0013	
Y-91	0.099	0.047	0.0161	
Zr-95	21.7	13.8	5.52	
Nb _m	0.0557	0.0309	0.0124	
Nb	19.3	19.3	11	
Mo-99	0.0184	small	small	
Te _m	0.0008	small	small	
Ru-103	19.9	7.3	1.6	
Rh _m	small	small	small	
Rh-105	small	small	small	
Ru-106	0	0	0	no γ
Rh	1.54	1.34	1.15	
Ag-110 _m	0.0055	0.00433	0.0038	
Ag-111	0.0411	0.00015	small	
Cd-115	small	small	small	
In _m	small	small	small	
Sn-119 _m	small	small	small	
Sn-121	0	0	0	no γ
Sb-122	small	small	small	
Sn-123 _m	0	0	0	no γ
Sn	small	small	small	
Sn-125 _m	0.0625	0.0007	small	
Sb	0.0233	0.0233	0.0233	
Te _m	small	small	small	
Sb-127	0.23	small	small	
Te _m	small	small	small	
Te	0	small	small	no γ
Te-129 _m	0.0393	0.0111	0.0016	
Te	1.8	0.525	0.21	
Te-131 _m	small	small	small	
Te	small	small	small	
I	4.61	0.027	small	
Xe _m	0.155	0.0078	0.000065	
Te-132	0.093	small	small	

TABLE IV (Continued)

<u>Isotope</u>	<u>30-day r/hr</u>	<u>90-day r/hr</u>	<u>180-day r/hr</u>	<u>Remarks</u>
I	1.28	small	small	
Xe-133 _m	0.00046	small	small	
Xe	small	small	small	
Cs-134	0.00053	0.00049	0.00046	
Cs-136	0.781	0.0026	0.00026	
Cs-137	0	0	0	no γ
Ba _m	0.211	0.211	0.211	
Ba-140	7.26	0.265	0.00196	
La	139	5.27	0.0377	
Ce-141	0.69	0.202	0.0274	
Ce-143	small	small	small	
Pr	0	0	0	no γ
Ce-144	0.0188	0.0157	0.0139	
Pr	0.44	0.371	0.324	
Nd-147	0.223	0.00576	0.00002	
Pm	0	0	0	no γ
Pm-148	small	small	small	
Pm-149	small	small	small	
Pm-151	small	small	small	
Sm	small	small	small	
Eu-152	small	small	small	
Eu-153	0.00008	small	small	
Eu-154	small	small	small	
Eu-155	0.00168	0.00151	0.00142	
Eu-156	2.4	0.163	0.00261	
Tb-160	0	0	0	no γ
Tb-161	<u>0.00053</u>	<u>small</u>	<u>small</u>	
Total	222	49	20.2	

TABLE V

FOURTEEN-INCH PRIMARY PIPING SURFACE DOSE RATE

Isotope	30-day mr/hr	90-day mr/hr	180-day mr/hr	Remarks
An-72	small	small	small	
Ga	small	small	small	
As-76	small	small	small	
As-77	0	0	0	no γ
Se _m	small	small	small	
Se-79	0	0	0	no γ
Br-82	small	small	small	
Kr-85	small	small	small	
Rb-86	small	small	small	
Sr-89	0	0	0	no γ
Sr-90	0	0	0	no γ
Y	1.05(10) ⁻³	0.00105	0.00105	
Y-91	0.87	0.413	0.142	
Zr-95	191	121	48.6	
Nb _m	0.49	0.272	0.109	
Nb	170	170	97	
Mo-99	0.162	small	small	
Te _m	small	small	small	
Ru-103	175	64.2	14.1	
Rh _m	small	small	small	
Rh-105	small	small	small	
Ru-106	0	0	0	no γ
Rh	13.5	11.8	10.1	
Ag-110 _m	0.05	0.04	0.03	
Ag-111	0.36	small	small	
Cd-115	small	small	small	
In _m	small	small	small	
Sn-119 _m	small	small	small	
Sn-121	0	0	0	no γ
Sb-122	small	small	small	
Sn-123 _m	0	0	0	no γ
Sn	small	small	small	
Sn-125 _m	0.55	small	small	
Sb	0.205	0.205	0.205	
Te _m	small	small	small	
Sb-127	2.02	small	small	
Te _m	small	small	small	
Te	0	0	0	no γ
Te-129 _m	0.34	0.097	0.014	
Te	15.8	4.62	1.85	
Te-131 _m	small	small	small	
Te	small	small	small	
I	40.5	2.38	small	

TABLE V (Continued)

<u>Isotope</u>	<u>30-day mr/hr</u>	<u>90-day mr/hr</u>	<u>180-day mr/hr</u>	<u>Remarks</u>
Xe _m	1.35	0.53	small	
Te-132	0.82	small	small	
I	11.4	small	small	
Xe-133 _m	small	small	small	
Xe	small	small	small	
Cs-134	0.00466	0.0043	0.0041	
Cs-136	6.86	0.023	0.0023	
Cs-137	0	0	0	no γ
Ba _m	1.86	1.86	1.86	
Ba-140	64	2.33	0.0173	
La	1220	46.3	0.332	
Ce-141	6.1	1.77	0.241	
Ce-143	small	small	small	
Pr	0	0	0	no γ
Ce-144	0.165	0.138	0.122	
Pr	3.87	3.26	2.85	
Nd-147	1.96	0.507	0.000202	
Pm	0	0	0	no γ
Pm-148	small	small	small	
Pm-149	small	small	small	
Pm-151	small	small	small	
Sm	small	small	small	
Eu-152	small	small	small	
Sm-153	small	small	small	
Eu-154	small	small	small	
Eu-155	0.0148	0.0133	0.0125	
Eu-156	21.3	1.44	0.023	
Tb-160	0	0	0	no γ
Total	1950	431	178	

ANALYSIS

Fission product information from reference (1) was used in the calculations. Reference (1) gives the saturation activities of the various important fission products as a function of the neutron flux, the percent of saturation as a function of exposure, and the decay as a function of time after shutdown. These curves consider parent-daughter transients as well as removal by neutron absorption. Reference (1) also supplied gamma information for each isotope.

The curves in Reference (1) were developed for U-235 fission products only. Since the rupture occurred in a plutonium element, an extrapolation for plutonium yields was necessary. This was done by multiplying the results by the Pu-239/U-235 fission product yield ratio for each isotope. The yield information was obtained from the November, 1960, issue of Nucleonics. This means of extrapolation, while academically correct, should give sufficiently accurate data for the purpose.

The parameters used in the calculation were the following:

Time of irradiation: $\frac{7.47(10)^{-3} \text{ KW-days exposure}}{1025 \text{ KW}} = 7.3 \text{ days}$

Neutron flux: Calculated at about $6(10)^{13}$ nv in the region of the rupture.

Decay times were taken at 10 days, 30 days, 90 days, 180 days.

Radiation levels were calculated for three situations. The first assumed the fission products concentrated in a point source surrounded by one inch of steel shielding. Radiation levels were calculated for a distance of one foot, using an expression given by Van Tuyl:⁽²⁾

$$D_1 = \frac{6 \text{ EC Be}^{-\mu t}}{R^2}$$

where D_1 is the dose rate in r/hr, E is the gamma energy in Mev, C is the number of gamma curies, B is the attenuation buildup factor, $e^{-\mu t}$ is the attenuation factor, and R is the distance from the point source in feet. Van Tuyl gives the quantity $\text{Be}^{-\mu t}$ for various materials (including steel) as a function of gamma energy and thickness. His values were used in these calculations.

(1) Blomeke, J. O., and Mary F. Todd, Uranium-235 Fission Product Production as a Function of Thermal Neutron Flux, Irradiation Time, and Decay Time. ORNL-2127, Part 1, Volume 1, Revised November, 1958, Unclassified.

(2) Van Tuyl, H. H., Fission Product Radiation and Shielding Calculations, May 16, 1961, HW-69533, Unclassified.

It should be emphasized in using the above formula that C is the gamma curies and may not be the same as the number of curies of the isotope. An example is Rb-86 in which only 12% of the disintegrations are accompanied by a gamma. In this case, 100 curies of Rb-86 would only be 12 gamma curies.

The second situation was the same as the first except that the iron shielding was removed. This means simply that $Be^{-\mu t}$ is unity.

The third situation assumed the fission products dispersed uniformly throughout the primary coolant system. The surface dose rates for the 14-inch primary piping, assuming a one-inch thickness of steel, was calculated.

There is no mathematically rigorous way to calculate the dose rate at the side of a cylindrical source. When the ratio of the distance from the cylinder axis to the detector to the cylinder radius is large, a line source is often used for an approximation. A different method was used to calculate the surface dose rate in the piping, and the results compared favorably with results calculated by the line source method.

Knowing that the specific activity is C/V curies per unit volume of coolant, where V is the volume of coolant (cm^3), the total activity per unit length of pipe is $\pi r^2 C/V$ where r is in cm. The gammas produced will, by symmetry of the pipe cross section, pass through the piping in a uniform gamma flux. The area per unit length of pipe which the gammas cross is $2\pi r$. Therefore, the gamma flux is $\frac{\pi r^2 C/V}{2\pi r}$ gamma curies per cm^2 . The dose rate equation given earlier may be written:

$$D_1 = \frac{4\pi \text{ 6EC } Be^{-\mu t} (30.5)^2}{4\pi r^2}$$

where $(30.5)^2$ has been introduced to change r to units of cm. In the above term, the quantity $C/4\pi r^2$ represents the unshielded gamma flux from the point source. Trading the gamma flux terms for the two situations, this expression may be altered to give the piping dose rates. This gives:

$$D_2 = 4\pi \text{ 6E } (30.5)^2 (rC/2V) Be^{-\mu t}$$

or simply
$$D_2 = 8.8(10)^{-3} D_1$$

Thus the surface dose rates for the primary piping are simply $8.8(10)^{-3}$ times the dose rate for the shielded point source.

J. A. Stoddard
PRTR Technical Planning

JA Stoddard:dde

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