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TITLE

Tentative Estimates of Radioactivity Associated With Recycle Plutonium

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TENTATIVE ESTIMATES OF RADIOACTIVITY ASSOCIATED WITH RECYCLE PLUTONIUM

The following information has been developed at your request in order to estimate the amount of radioactivity associated with the plutonium which is to be recycled in the Plutonium Recycle Program Reactor (PRPR). The numbers are necessarily qualitative and require experimental verification before application.

Conclusions

As a basis of comparison with the estimates presented below, the plutonium presently processed in 234-5 has a surface dose rate of about 1.5 r/hr of gamma from bare metal shapes or about 350 mr/hr of gamma from coated shapes. Neutron emission from the plutonium fluoride station represents about 1 rem/hr.

The conclusions as presented refer to plutonium isotopic compositions derived from the PRPR that give the highest value for the specific activity considered.

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(1) Alpha Radiation: It is predicted that there will be a slight increase in alpha radiation (perhaps a factor of 1.5 times our present plutonium). This occurs when the recycle plutonium has a high concentration of Pu-240.

(2) Beta Radiation: There is a very large increase in the number of beta particles over presently produced plutonium due to the increased percentage of Pu-241. Fortunately, their very low energy (0.02 MEV) will require little in the way of shielding.

(3) Gamma Radiation: The estimated values for gamma radiation are far from precise due both to their complex energy spectrum and to the lack of experimental data. It is therefore recommended that further experimental measurements be made with particular emphasis on gamma radiation. These measurements preferably should include shield thickness as a variable since the gamma energy spectrum of recycle plutonium differs from that of Pu-239.

If freshly separated (Am-241 negligible), it is predicted that unshielded recycle plutonium will show little increase in gamma over unshielded Pu-239. For recycle plutonium which is only slightly shielded there should be a substantial increase (say a factor of 5) in gamma radiation over slightly shielded Pu-239 due to 0.1, 0.147, (Pu-241) and 0.06 (Am-241) MEV gammas. The reason for this apparent anomaly in the case of light shielding is that the 0.017 MEV x-rays which form a large fraction of the total gamma from Pu-239 will attenuate rapidly. As plutonium is allowed to age after separation, the contained Pu-241 isotope decays to Am-241. After a period of six months it is predicted that the high output of 0.06 MEV gamma derived from Am-241 will raise the unshielded gamma emission of recycle plutonium, 2 to 5 times greater than Pu-239.

(4) Neutron Radiation: The number of neutrons produced by α -n reactions in recycle plutonium increases a factor of 2 over those from Pu-239. In addition, neutrons from the spontaneous fissions of Pu-240 and Pu-242 in recycle plutonium are predicted to cause a surface dosage rate of about 1 rem/hr. from an unmoderated mass such as the present shape.

Discussion

Elements & Isotopes Considered

Calculations have been made relating the amounts of Pu-239, Pu-240, Pu-241, and Pu-242 with exposure for the PRPR. In summary, the isotopic composition ranges from nearly pure Pu-239 to nearly pure Pu-242. Somewhere near the middle of the exposure period, (300 to 600 days) the four isotopes are present in approximately equal quantities. For the cases considered, Pu-240 (or Pu-241) is always less than 50% of the total plutonium. Plutonium-238 was not calculated, but the few available analytical results show it to be present up to a

maximum of about 0.2 weight per cent. Plutonium-243 will be virtually absent from plutonium "cooled" for even two days because of its short (5 hr.) half-life.

The two americium isotopes, 241 and 243, are presumably removed during chemical processing. Am-243 will not grow in after processing since the mother element, Pu-243, will not be present due to decay during "cooling" prior to the separations step. Am-241 will, however, grow in with time from Pu-241. For example, at six months after processing, 0.026 atoms of Am-241 will be present for each atom of Pu-241 originally present. The amount of Am-241 at any time t, after separation, is found from the following equation:

$$\text{Am-241} = 1.028\ (\text{Pu-241})(e^{-0.00147t} - e^{-0.0533t})$$

where Am-241 = no. of Am-241 atoms at time t
Pu-241 = no. of Pu-241 atoms at time o
t = time since separation, yrs.

The quantity of radioactivity which results from fission products is of course dependent upon the specific separations process employed, and will be in addition to the quantities computed here.

Specific Activity of Recycle Plutonium

The specific activities for the isotopes of interest are given in Table I, below in terms of events/min/mg.

TABLE I

Isotope	Half-Life yrs.	Alpha α/min/mg	Beta β/min/mg	Gamma γ/min/mg	Neutrons: Spont Fiss. n/min/mg	Neutrons: α-n(1) n/min/mg.F	Neutrons: α-n(2) n/min/mg.Pu
Pu-238	90	3.7×10^{10}	0	1.2×10^{7}	185	2.6×10^{5}	4000
Pu-239	24,300	1.4×10^{8}	0	2.5×10^{4}(3)	0.0016	720	12
Pu-240	6600	5.0×10^{8}	-	2.5×10^{4}	75	2900	48
Pu-241	13	2.5×10^{6}	2.5×10^{11}	1.0×10^{6}	-	41	0.2
Pu-242	3.8×10^{5}	8.6×10^{6}	0	0	125	37	0.6
Am-241	470	7×10^{9}	0	2.8×10^{9}(4)	-	4.8×10^{4}	-

(1) Computed for a small percentage of impurity (in this case fluorine) in plutonium metal. The yield of neutrons from fluorine is on the order of 10 higher than from other common impurities*.

(2) Computed for a small percentage of plutonium in a diluent, in this case carbon; other diluents have lower or higher yields.*

(3) Excludes 17 KEV x-rays.

(4) Number of 60 KEV gammas, there are an additional unknown number of lesser energy.

* See Table 1.1.1, p. 2, Reactor Handbook, RH-1 (AECD 3645).

An inspection of the table leads to the following conclusions:

(1) Alpha activity of recycled plutonium is expected to be only slightly higher (say a factor of 1.5) than our present plutonium (Pu-238 and Am-241 are present in such small percentages that they make only a small contribution to the total).

(2) Neutrons from α-n reactions may be expected to double over those presently found. (Pu-240 has about 4X the number of α-n's, but is present to a maximum concentration of less than 50 wt %.)

(3) Beta particles although present in large numbers from Pu-241 should not present a serious problem because of their very low energy (0.02 MEV).

The gamma ray problem requires a more thorough inspection because of the several energies associated with them. Table II shows the calculated gamma radiation for the isotopes of interest.

TABLE II

Isotope	Energy of Gamma Ray MEV	R/hr/g @ 1 cm	R/hr. "Semi Infinite" Slab
Pu-238	0.045	3.2	2.2
Pu-239	0.017	5-10	~1.5
	0.039	2.84×10^{-4}	1.8×10^{-4}
	0.053	2.19×10^{-3}	2.1×10^{-3}
	0.100	1.73×10^{-3}	5.6×10^{-3}
	0.127	1.08×10^{-3}	5.2×10^{-3}
	0.384	2.26×10^{-3}	3.2×10^{-2}
Pu-240	0.044	6.65×10^{-3}	4.6×10^{-3}
Pu-241	0.100	0.21	0.68
	0.147	0.065	0.35
Am-241	0.06	606(1)	~700
	0.06	16(2)	~18

(1) 606 R/hr./g. of Am-241
(2) 16 R/hr./g. of initial Pu-241 aged 6 mo. to form Am-241

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The column headed R/hr./g. is a conversion from Table I and represents the total emission of the specific isotopes with no allowance for self-absorption. The column headed R/hr. results from a rough estimation of the dose rate from the surface of a piece of plutonium metal.

Bare plutonium metal as presently handled in 234-5 produces a surface dose rate of about 1.5 R/hr, while coated pieces produce about 350 mr/hr. The majority of this gamma as measured is attributed to the 17 KEV x-rays from Pu-239. There is a possibility that some of the other isotopes may also emit such x-rays, and in this event, the surface dosage rate of that particular isotope will be higher than shown in Table II.

Based on the estimated values presented in Table II, no appreciable increase in gamma is expected for bare recycle plutonium metal until the contribution of Am-241 becomes apparent. At six months time after separations and with a considerable amount of Pu-241 originally present, the gamma from Am-241 may reach a level 2-5 times the gamma from Pu-239. The gamma levels for very lightly shielded recycle plutonium will be somewhat higher than for Pu-239 because of increased amounts of 0.06, 0.1, and 0.147 MEV gamma.

Neutron emission will be higher in recycle plutonium. Two sources of neutron emission are considered. The predicted number of neutrons from alpha-neutron reactions will be about double those now experienced. These neutrons will result from Pu-240 which has about 4 times the number of α -n events (Table I, compare Pu-239 with Pu-240), but which reaches a maximum concentration of about 50 per cent of the total plutonium. The other source of neutrons will be the spontaneous fissions of Pu-240 and Pu-242. Preliminary calculation for the case when Pu-242 forms a large fraction of the total recycle plutonium indicates a neutron dosage rate of about 1 rem/hr. from an unmoderated mass such as the present shapes.

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