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RADIATION DENSITIES AND TBP  
RADIOLYSIS DURING THOREX  
SHORT DECAY RUNS

W. Davis, Jr.

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CHEMICAL TECHNOLOGY DIVISION

Chemical Development Section B

RADIATION DENSITIES AND TBP RADIOLYSIS DURING  
THOREX SHORT DECAY RUNS

Wallace Davis, Jr.

DATE ISSUED

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OAK RIDGE NATIONAL LABORATORY  
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ABSTRACT

A general equation was derived to calculate the total radiation dose received by the solvent in a pulsed column, assuming ideal mixing of organic and aqueous phases. With this equation and data from the Thorex short-decay runs, radiation dose densities and the amount of TBP that might be decomposed by radiation in the first cycle extraction column were calculated. The average radiation densities were 0.13-0.24 watt-hr/liter, which would decompose 0.01% of the TBP in the 40% TBP--Amsco 125-82 extractant. The amount of acidic radiolysis products would probably be inadequate to precipitate thorium dibutyl phosphate in the 1A column at equilibrium, but would probably cause precipitation if the extractant residence time in the extraction section was increased by a factor of 3 over the 1.2-1.5 min that prevailed during the short-decay runs.

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## 1.0 INTRODUCTION

This report gives the results of a detailed examination of the radiation density and tributyl phosphate, TBP, decomposition by radiation in the Thorex short-decay runs. The mathematical methods presented here are applicable to analyses of effects of homogeneously distributed radiation sources, such as fission products, in general; however, only the effects of Pa-233 are analyzed in this report.

The highest radiation dose densities encountered during radiochemical reprocessing of nuclear fuels at ORNL are those of the Thorex Pilot Plant short-decay runs. During the first of these runs, SD-1, precipitation of a thorium compound, probably thorium dibutyl phosphate, in the first cycle extraction column (1A) caused plugging of the LAP line, containing the product-bearing extractant from this column. In addition, gel formation in the second cycle stripping column necessitated shutdown on a 3- to 5-day cycle instead of the 15-day cycle that obtained for the Thorex long-decay runs. Thus, the radiation level in the first cycle affected the second-cycle operations. Data obtained during the Thorex short-decay runs indicate that the radiation levels in the 1A column were close to those that could cause serious trouble. In run SD-1 the extractant residence time in the 1A column was greatly increased, increasing the exposure. When the column was operating at equilibrium there were no major difficulties.

Several members of the Chemical Technology Division were most helpful in providing general information and specific data concerning the Thorex Pilot Plant. The author particularly wishes to thank W. T. McDuffee, R. H. Rainey, A. T. Gresky, E. M. Shank, and J. R. Parrott.

## 2.0 RADIATION DOSE DENSITY

In all solvent extraction processes, including the Thorex process, more than 99% of the radioactive nuclides in the feed stream are separated from the fissile or fertile materials in the first cycle extraction column. Therefore more than 99% of any solvent degradation due to radiation occurs in this first column and we may neglect radiation degradation in other process equipment.

The first cycle extraction column of the Thorex process is a pulsed column in which the section below the feed point is designated as the "extraction" section and that above the feed point as the "scrubbing" section. Pertinent factors concerning this column are:<sup>1,9</sup>

Inner diameter of column, 5.047 in. (5 in. IPS)

Distance between similar surfaces of adjacent pulse plates, 2 in.

Thickness of pulse plates, 1/32 in.

- $v$  = geometrical volume contained between the lower surface of plate  $i$  and the lower surface of plate  $i-1$ , 0.643 liter with a few minor exceptions<sup>1</sup>
- $v_i^o$  = volume of organic phase contained in stage  $i$ , between plates  $i-1$  and  $i$ , liters
- $f_i^o$  =  $v_i^o/v$
- $n$  = number of geometrical stages in the column, 182, where the volume between two adjacent plates constitutes one stage
- $n_f$  = number of the feed stage, 55, during the short-decay runs
- $Q$  = extractant flow rate, liters/hr
- $F$  = feed flow rate, liters/hr
- $S$  = scrub flow rate, liters/hr
- $\tau$  = total operating time with radioactive feed, hr
- $P_i^o$  = specific power absorbed by the organic phase in stage  $i$ , watts/liter
- $E_i^o$  = energy absorbed by the solvent in stage  $i$ , watt-hr
- $D$  = average dose density, watt-hr/liter, received by the extractant during the time  $\tau$
- $I$  = total volume of extractant used during the time  $\tau$ , liters
- $r$  = pulse rate, pulses/hr
- $t$  = residence time of extractant in that portion of the column having significant radiation power density, hr
- $N$  = number of extractant inventory passes through the first cycle during operation with radioactive feed
- $C_{ip}^a$  = concentration of Pa-233 in the aqueous phase in stage  $i$ , g/liter
- $C_{if}^a$  = concentration of (other) radioactive nuclide  $f$  in the aqueous phase of stage  $i$ , g/liter

- $C_{if}^0$  = concentration of (other) radioactive nuclide f in the organic phase of stage i, g/liter
- $W_P$  = specific power of  $\beta$  + internal conversion electron ( $e^-$ ) +  $\gamma$  radiation from Pa-233 in the 1A column, 36.6 watts/g\*
- $W_f$  = specific  $\beta$ ,  $e^-$ , or  $\gamma$  power from nuclide f which is attenuated by the liquid in the column, watts/g

In calculating the average radiation dose absorbed by the extractant, it is convenient to consider the problem in terms of the following steps:

1. The quantity  $v_i^0$  of extractant between plates i-1 and i is irradiated at the equilibrium power level  $P_i^0$  for the total time  $\tau$  during which radioactive solution is being fed. From this, the contribution of radiation in stage i to the total radiation energy absorbed is

$$E_i^0 = \tau v_i^0 P_i^0, \text{ watt-hr} \quad (1)$$

2. The contributions of radiation in all stages are added to give the total energy absorbed:

$$\sum_{i=1}^n E_i^0 = \tau \sum_{i=1}^n v_i^0 P_i^0 \quad (2)$$

3. The average dose density, D, is obtained by dividing the total energy absorbed by the total extractant inventory, I:

$$D = \frac{1}{I} \sum_{i=1}^n E_i^0 = \frac{\tau}{I} \sum_{i=1}^n v_i^0 P_i^0 = \frac{\tau v}{I} \sum_{i=1}^n f_i^0 P_i^0 \quad (3)$$

Finally, from the definitions listed above, the power density,  $P_i^0$ , in stage i can be expressed in terms of radioactive nuclide concentrations and specific powers if it is assumed that aqueous and organic phases are completely dispersed:

---

\* See Appendix for the derivation of this value.

$$P_i^O = \left[ C_{ip}^a W_p + \sum_f C_{if}^a W_f \right] \frac{v-v_1^O}{v} + \frac{v_i^O}{v} \sum_f C_{if}^O W_f \quad (4)$$

Equation 4 contains a term for radiation from Pa-233 dissolved in the aqueous phase but none for this element dissolved in the organic phase since the distribution coefficient (o/a) is of the order of  $10^{-3}$ . Although a term for radiation due to fission products dissolved in the organic phase is included in eq. 4, this term is also very small for the Thorex short-decay runs and will be neglected in later calculations.

The quantities required to calculate the radiation dose density received by the extractant in the 1A pulsed column are given in eqs. 3 and 4. Of these,  $\tau$ ,  $v$ , and  $I$  are readily determined experimentally (Table 1 and definitions), but  $P_i^O$  and  $v_i^O$  or  $f_i^O$  must be calculated from other quantities. These calculations are outlined in the next two sections.

### 2.1 Radiation Power Density

About 98% of the total radiation in the Thorex short-decay runs was due to Pa-233.<sup>9</sup> This nuclide does not reflux significantly since its distribution coefficients<sup>6</sup> (o/a) are of the order of  $10^{-3}$ . Therefore the Pa-233 concentration should be essentially constant in the extraction section of the 1A column and equal to  $FC_p^{LAF}/(F+S)$  (Fig. 1). The aqueous Pa-233 concentration was measured only at the feed tank and at a hold tank in the 1AW discharge line. The ratio  $C_p^{LAW}/C_p^{LAF}$  varied between 0.39 and 0.77 (Table 1, excluding the value 1.30<sup>9</sup>); the average ratio of 0.60 compares favorably with a calculated ratio of 0.555 for the condition of no reflux, although this agreement does not preclude reflux per se. Experimental values of  $C_p^{LAW}$  were used in Table 1 for the quantity  $C_{ip}^a$  of eq. 4. Finally, although the radioactive rare earth element reflux<sup>6</sup> will introduce an error of 10 to 20% in the determination of  $P_i^O$ , the concentrations  $C_{if}^O$  of eq. 4 were assumed equal to zero in this report.

### 2.2 Extractant Holdup

The extractant holdup,  $v_i^O$ , or its equivalent,  $f_i^O$ , cannot be calculated, nor has it been measured, for each geometrical stage. Only an

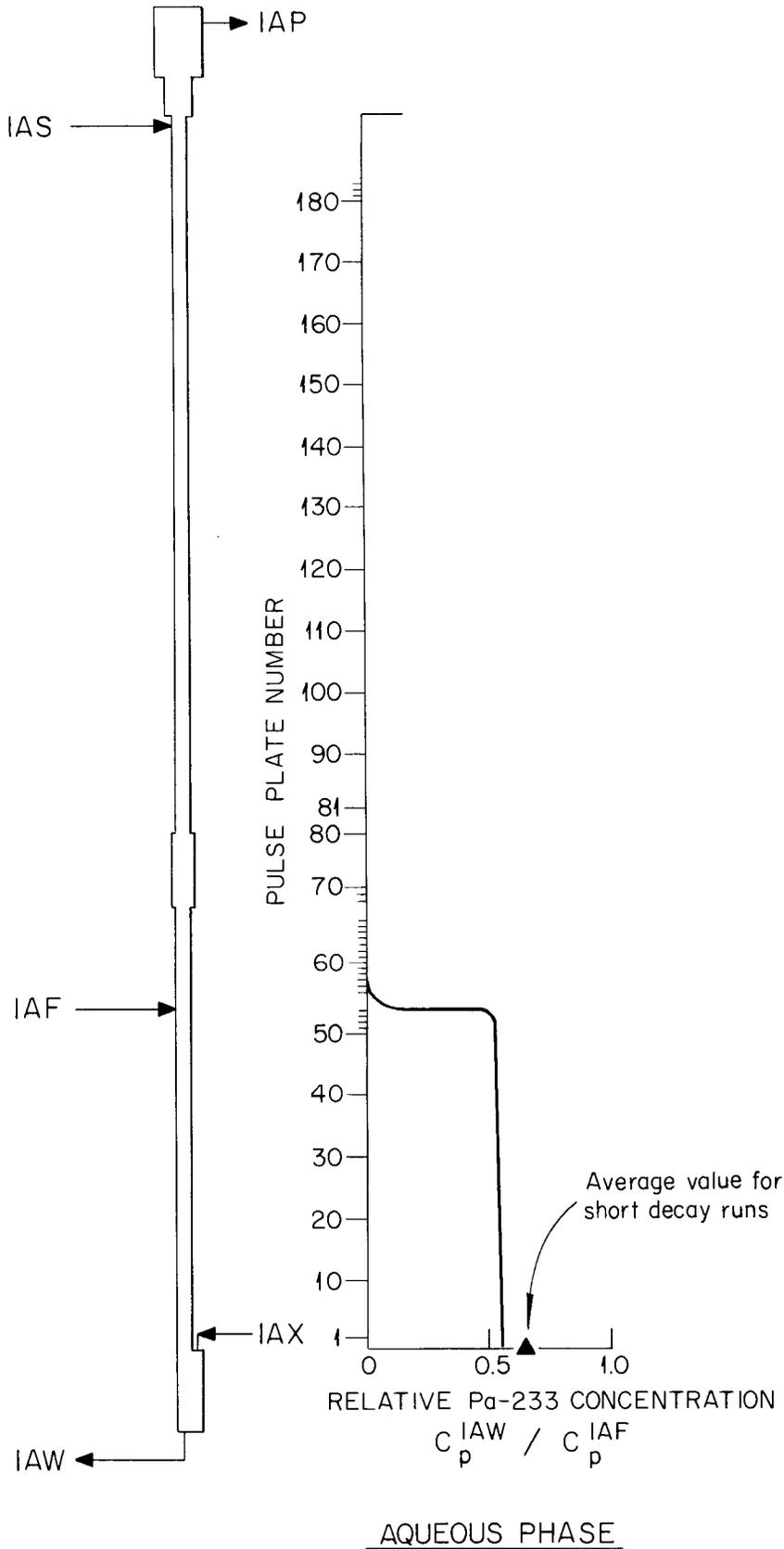


Fig. 1. Approximate Pa-233 Profile in IA Column.

average value can be calculated. This may be done by use of the data and correlation of McNamee, Rogers, and Dinsmore,<sup>10</sup> eq. 5, where  $\bar{f}^0$  is the average fraction of the column volume occupied by extractant during operation,  $\bar{f}^{0*}$  the corresponding value, 0.15 (ref. 10), at the onset of column flooding, and  $(Q+F+S)/(Q+F+S)^*$  is the relative approach to flooding, 0.30 (ref. 9) in the short decay runs:

$$\bar{f}^0 = \frac{(Q+F+S)}{(Q+F+S)^*} \bar{f}^{0*} = \frac{\bar{v}_i^0}{v} = 0.045 \quad (5)$$

The total extractant flow rate,  $Q$ , may be equated to the pulse frequency times the extractant flow per pulse,  $k_i v_i^0$ :

$$Q = k_i v_i^0 r = \frac{NI}{\tau} \quad (6)$$

By dividing eq. 6 by  $vr$  and assuming that all the extractant between two plates moves up one geometrical plate at every pulse, i.e., that  $k_i = 1$  plate per pulse, we can obtain a second estimate of the average fractional volume,  $\bar{f}^0$ , occupied by the extractant (Table 1):

$$\frac{\bar{v}_i^0}{v} = \bar{f}^0 = \frac{Q}{vr} \quad (7)$$

### 2.3 Numerical Results

Except in the first short-decay run, Pa-233 appears not to have refluxed and not to have precipitated in the 1A column. Therefore in runs SD-2 and SD-3, the Pa-233 in the extraction section of the 1A column may be set equal to its concentration in the LAW stream. Because there appears to have been no reflux of Pa-233, the radiation level above the feed plate can be considered negligible, in which case the upper limit,  $n$ , of summation in eqs. 2 and 3 is 55 rather than 182 plates. This concept is summarized as the Pa-233 profile (Fig. 1). On this basis, the average dose received by the extractant, 40% TBP in Amsco 125-82, in the Thorex short-decay runs was 0.13 to 0.24 watt-hr/liter; single-pass dose densities varied between 0.05 and 0.12 watt-hr/liter except in the

Table 1

	Average Values			
	Run SD-1 <sup>a</sup>	Run SD-2	Run SD-3	
Pa-233 conc in LAW, g/liter	0.18	0.068	0.175	
(Pa-233 conc in LAW)/(Pa-233 conc in LAF)	0.53	0.62	0.63	
Pulse frequency, r, pulses/hr	2340	2400	2340	
LA operation time with radioactive feed, $\tau$ , hr	114	241	113	
Solvent inventory, I, liters	5500	6940	5200	
Total solvent pumped, liters	28,200	36,850	7750	
Total operating time, hr	300	428	88	
Average extractant flow rate, Q, liters/hr	94	86	88	
No. of solvent passes during operation with radioactive feed, N	1.95	2.99	1.91	
Extractant residence time	eq. 9	0.0190	0.0207	0.0202
in extraction section, t, hr	eq. 10	0.0235	0.0229	0.0235
Pa-233 radiation power in LAW, watts/liter	6.6	2.5	6.4	
Pa-233 power in LA extraction section, $\overline{P^O} = \overline{P^A}$ , watts/liter	6.2	2.3	6.1	
Radiation dose per pass through LA, watt-hr/liter	0.12	0.048	0.12	
Total dose, <sup>b</sup> D, watt-hr/liter	eq. 3	0.23	0.15	0.24
	eq. 8	0.20	0.13	0.21
Extractant volume fraction, $\overline{f^O}$	eq. 5	0.045	0.044	0.044
	eq. 7	0.062	0.056	0.058

<sup>a</sup>Values for run SD-1 listed in this column do not apply to that portion of the run after precipitation of Pa-233. Such precipitation may have increased the radiation dose density by a factor of 10.

<sup>b</sup>In both cases the value  $\overline{f^O} = 0.045$  was used.

SD-1 run (Table 1). These numbers may be uncertain by as much as 50%, primarily because of two factors: (1) the two methods for calculating the extractant holdup (eqs. 5 and 7 and Table 1) give results that differ by nearly 30%, i.e., 0.045 vs 0.058 as the two values calculated for the fraction of the column volume occupied by the extractant; (2) reflux of the rare earth elements, which might contribute 10 to 20% of the total radiation, has been neglected.

Values of dose density presented in this paper are lower than those of McDuffee and Yarbrow<sup>9</sup> by a factor of about 20. Reasons for this difference are outlined as follows:

1. The organic holdup reported here is 4.5 to 5.8% of the column volume, the first value being based on more direct experimental data, as contrasted with an assumed value of 20% used by McDuffee and Yarbrow. This item contributes a factor of 4.45.

2. The radiation density is equal to the fraction  $(v-v^0)/v = 0.95$  times the Pa-233 concentration,  $C_p^{1AW}$ , in the 1AW stream, not to the concentration  $C_p^{1AW}$  itself. This item contributes a factor of 1.05.

3. McDuffee and Yarbrow assumed that all the extractant in the 1A column was subject to the same radiation density as that in the extraction section of this column. Such an assumption is not warranted since distribution coefficient data<sup>6</sup> show that Pa-233 will not reflux very much. Thus only the lower  $\sim 55/182$  of the extractant in the column actually received the full dose (see Fig. 1 for estimated profile). This item contributes a factor of about 3.3 to the factor 20.

4. On the basis of recent analyses<sup>2,11</sup> of the  $\beta$ , internal conversion electron, and  $\gamma$ -ray emission from Pa-233, the specific power of this radiation in the 1A column is only 36.6 watts/g rather than the value 50 watts/g used by McDuffee and Yarbrow. This item contributes a factor of 1.33.

#### 2.4 Alternative Calculations

A relatively large number of experimental variables and derived quantities are involved in calculating the density of the radiation in a pulsed column. Some of these can be used in different combinations.

For example, the average power density times the residence time,  $t$ , of extractant in the extraction section of the 1A column is the average dose density per pass,  $D_1$ , and the total dose is then  $D_1 N$ , where  $N$  is the number of inventory passes:

$$D_1 = t \overline{P^0}$$
$$D = N D_1 = N t \overline{P^0} \quad (8)$$

The quantity  $t$  may be estimated two ways, just as  $\overline{P^0}$  was estimated two ways (eqs. 5 and 7):

$$t \approx \frac{(Q+F+S)}{(Q+F+S)^*} \frac{\overline{P^0}^*}{\overline{P^0}} \cdot \frac{V}{Q} \frac{n_f}{n} \quad (9)$$

or

$$t \approx n_f / r \quad (10)$$

Power densities during the Thorex short-decay runs (Table 1) were of the order of 2 to 6 watts/liter, extractant residence times in the extraction section were about 0.02 hr, and dose densities per pass 0.05 to 0.12 watt-hr/liter.

### 2.5 Extent of Decomposition of TBP by Radiation

The decomposition of TBP in Amsco 125-82 solutions has been studied extensively enough that the weight fraction,  $W_{acid}$ , of MBPA and DBPA produced in a solution containing  $W_{TBP}$  weight fraction TBP and exposed to radiation at the level  $D$  watt-hr/liter can be accurately expressed:<sup>3</sup>

$$W_{acid} = 9.20 \times 10^{-7} D W_{TBP} \frac{(\text{mol. wt. of acid})}{(\text{density of solution})} \quad (11)$$

Dibutyl phosphoric acid (DBPA) comprises about 85% of the acidic products and monobutyl phosphoric acid (MBPA) most of the residual 15%; hence, the molecular weight of the acid (eq. 11) is about 200 and the density of the solution is about 0.83 g/ml. If the radiolysis of TBP-Amsco solutions

was independent\* of  $\text{HNO}_3$  and the other solutes in the extraction section of the 1A column, the concentration,  $W_{\text{acid}}$ , of MBPA and DBPA formed by irradiation to the average of 0.2 watt-hr/liter level of the Thorex short-decay runs would be approximately  $2.9 \times 10^{-5}$  g per gram of solution, corresponding to 0.0029 wt % acids. To form these acids about  $3.8 \times 10^{-5}$  weight fraction of TBP was decomposed, i.e., 0.008%:

$$3.8 \times 10^{-5} \frac{\text{g TBP decomposed}}{\text{g solution}} \times \frac{100}{0.47 \text{ g TBP/g solution}} = 0.008\%$$

In terms of TBP losses, radiation decomposition is trivial; however, at a dose greater than about 0.4 watt-hr/liter, enough DBPA will be formed in a 40% TBP-Amsco solution to cause precipitation of thorium dibutyl phosphate\*\* in the extractant phase.<sup>3</sup> At present, the effects of nitric acid on the solubility of this compound in TBP-Amsco are not known. However, it is apparent that the 0.13 to 0.24 watt-hr/liter dose density of the Thorex short-decay runs is nearly great enough to cause precipitation of thorium dibutyl phosphate unless nitric acid causes a large increase in its solubility in the extractant. In run SD-3 (Table 1) the power density was 6.1 watts/liter, but since the average extractant residence time was only 0.02 hr (1.2 min), the average dose density per pass was only 0.12 watt-hr/liter. If the residence time was increased to 3 or 4 min or more by operational difficulties, precipitation of thorium dibutyl phosphate could be expected.

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\* More recent data<sup>13</sup> indicate that yields of DBPA and MBPA may be in the ratio 3:2 after contacting the TBP-Amsco with 2 M  $\text{HNO}_3$ . The total acid yield does not appear to have increased more than about 10% per mole of  $\text{HNO}_3$  in the aqueous phase.

\*\* Data obtained at Stanford Research Institute show the solubility of thorium dibutyl phosphate in any solution containing only TBP and Amsco to be less than 0.04 g/liter.

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4.0 APPENDIX

SPECIFIC POWER OF Pa-233 RADIATION

In the decay of Pa-233 to U-233, three partial spectra of  $\beta$  particles,<sup>2,11,12</sup> with maximum energies of 0.15, 0.257, and 0.568 Mev, are emitted (Table 2). By procedures outlined by Marinelli, Brinkerhoff, and Hine<sup>12</sup> or Dismuke, Rose, Perry, and Bell,<sup>4</sup> and the  $\bar{E}_\beta$  tables of the latter reference, average  $\beta$  energies and contributions of each group of  $\beta$  particles to the specific  $\beta$  power were calculated. The sum of the three latter terms is 6.86  $\beta$ -watts/g Pa-233. Since the maximum range of the maximum-energy  $\beta$  particles from Pa-233 is less than 0.07 in.<sup>7</sup> in Thorex 1A column solutions, which have densities in excess of that of water, it has been assumed in this report that all the  $\beta$  energy is attenuated by the solutions in this column.

Table 2. Specific  $\beta$  Power from Pa-233<sup>a</sup>

Maximum $\beta$ Energy, Mev	No. of $\beta$ Particles per 100 Pa-233 Decays	$(E_\beta)$ Av $(E_\beta)$ Max	Average $\beta$ Power, $\beta$ -watts/g Pa-233
0.15	37	0.273	0.756
0.257	58	0.276	5.062
0.568	5	0.299	1.045
			<u>6.863</u>

<sup>a</sup>The half-life used in these calculations was 27.0 d.

According to Brodie,<sup>2</sup> whose results are largely substantiated by Ong and Kramer,<sup>11</sup> about 95% of the U-233 atoms formed in the decay of Pa-233 are initially in excited states; deactivation to the ground state occurs through 13 transitions, most of which are combinations of  $\gamma$ -ray and internal conversion electron,  $e^-$ , emission processes (Table 3). The analysis of Brodie is important in the present work since energy of internal conversion electrons will be nearly completely absorbed by the solutions in a column such as the 1A, while a considerable portion of the  $\gamma$  energy will not be attenuated in this column. Specifically, the 20.8  $e^-$  watts/g Pa-233 (Table 3) will be essentially completely absorbed, while only about

8.9  $\gamma$ -watts/g Pa-233, i.e., 44% of the  $\gamma$  energy, will be absorbed in the 1A column (Table 4). It is apparent that if a piece of process equipment has dimensions in excess of 1 in., it is appropriate to assume that the  $\beta$  and  $e^-$  (internal conversion) energy is all absorbed; however, the Pa-233  $\gamma$  energy absorbed is dependent on equipment size until minimum dimensions are of the order of 18 in. or more. For the purpose of this report, the following specific power of Pa-233 has been used:

$\beta$ power	6.86	$\beta$ watts/g Pa-233
Internal conversion electron power	20.8	$e^-$ watts/g Pa-233
$\gamma$ energy in the 1A column	8.9	$\gamma$ watts/g Pa-233
Total	36.6	watts/g Pa-233

Table 3. Distribution of  $\gamma$ -ray and Internal Conversion Electron Radiation from Pa-233<sup>a</sup>

Transition No.	Energy of Transition, keV	Transition Intensity Distribution		Specific Power Distribution	
		$e^-$ per Decay of Pa-233	$\gamma$ per Decay of Pa-233	$e^-$ watts per g Pa-233	$\gamma$ watts per g Pa-233
1	17.2	b	b	--	--
2	28.7	0.04	0.02	0.141	0.071
3	40.7	0.02	0.01	0.100	0.050
4	58	0.02	0.01	0.143	0.071
5	76	0.075	0.035	0.701	0.327
6	87	0.06	0.05	0.642	0.535
7	104	0.04	0.03	0.512	0.384
8	272	-- Very weak --		--	--
9	301	0.02	0.01	0.741	0.741
10	313	0.43	0.37	16.560	14.249
11	342	0.03	0.04	1.262	1.683
12	400	--	0.023	--	1.132
13	417	--	0.023	--	1.180
Totals				20.80	20.42

<sup>a</sup>These are the assignments made by Brodie.<sup>2</sup>

<sup>b</sup>This transition has a total intensity of 5% or less, but a total energy contribution of only 0.105 watt/g Pa-233. Its contribution has been neglected because of the uncertainty of its assignment.

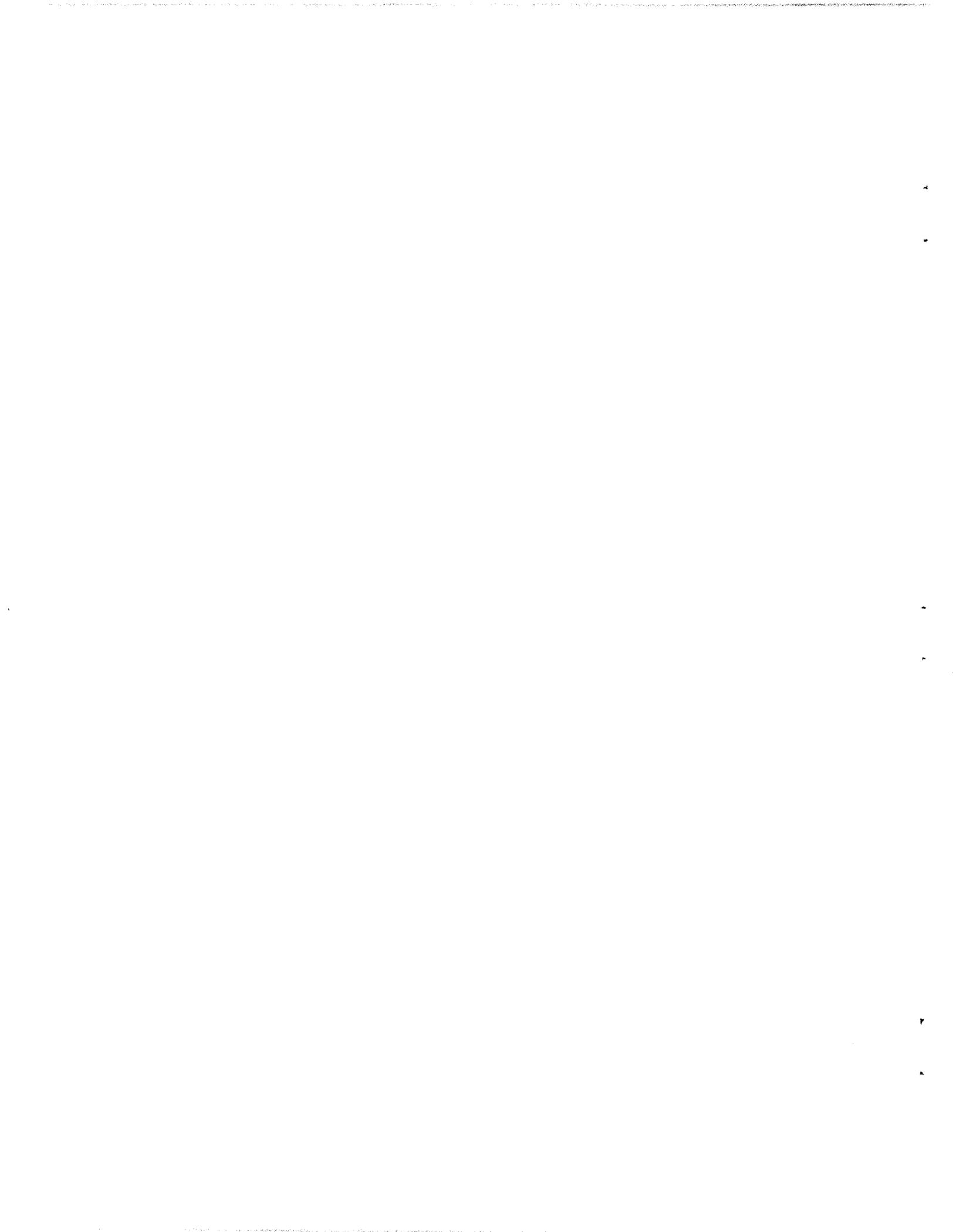
Table 4. Attenuation of  $\gamma$  Radiation From Pa-233

Transition No.	$\gamma$ -ray Energy, kev	Mass Attenuation Coefficient in Water, <sup>a</sup> $\mu_o/\rho$ , cm <sup>2</sup> /g	Linear Absorption Coefficient in 1A Column, <sup>b</sup> $\mu = \left(\frac{\mu_o}{\rho}\right) \bar{\rho}$ cm <sup>-1</sup>	Half Thickness for $\gamma$ -abs. in 1A Column, cm	Fraction of $\gamma$ Energy Absorbed in 1A Column <sup>c</sup>	$\gamma$ Energy Absorbed in 1A Column, $\gamma$ watts per g Pa-233
1	17.2	--	--	--	--	--
2	28.7	0.40	0.414	1.63	0.77	0.055
3	40.7	0.26	0.276	2.51	0.67	0.034
4	58	0.206	0.218	3.18	0.605	0.043
5	76	0.187	0.198	3.50	0.58	0.190
6	87	0.179	0.190	3.64	0.565	0.302
7	104	0.17	0.180	3.85	0.545	0.209
8	272	--	--	--	--	--
9	301	0.118	0.125	5.55	0.44	0.326
10	313	0.115	0.122	5.68	0.43	6.127
11	342	0.112	0.119	5.81	0.425	0.715
12	400	0.105	0.111	6.25	0.405	0.458
13	417	0.103	0.109	6.35	0.40	0.472
					Total	8.93

<sup>a</sup>Reference 7.

<sup>b</sup>The average density of solution in the extraction section of the 1A column,  $\bar{\rho}$ , is about 1.06 g/ml, based on 95% aqueous--5% organic holdup.

<sup>c</sup>Reference 5.



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