OAK RIDGE NATIONAL LABORATORY
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UNION CARBIDE CORPORATION
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U.S. ATOMIC ENERGY COMMISSION

ORNL-TM-686
COPY NO. - 1
DATE - September 27, 1963

LIMITING CRITICAL CONCENTRATIONS OF AQUEOUS NITRATE SOLUTIONS OF FISSILE AND FERTILE ISOTOPES

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ABSTRACT

Results of IBM-7090 MODRIC calculations of the limiting critical concentrations of aqueous nitrate solutions of U^{235}-U^{238}, U^{235}-Th^{232}, U^{233}-Th^{232}, Pu^{239}-U^{238}, and Pu^{239}-Th^{232} are presented.

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Limiting critical concentrations of aqueous nitrate solutions of mixtures of fissile and fertile isotopes were calculated. The calculations were made with the IBM-7090 MODRIC neutron diffusion code, using 34 group "finite concentration" cross sections, and techniques described in ORNL-3309.[1] Results are plotted in Fig. 1 and are tabulated in Tables 1 and 2.

This report supercedes ORNL CF 62-10-55[2] and includes data from that report.

The calculated limiting critical concentrations of fissile material are believed to be accurate within ±10% for enriched uranium of enrichment 3-100 wt %. The calculation overestimates the experimentally determined critical concentration of fully enriched uranium (12.05 ± 0.03 g U^{235}/liter)[3] by 3% and underestimates the experimentally determined critical concentration of 3.04% enriched uranium (20.2 ± 0.5 g U^{235}/liter)[4] by 10%. It is to be expected that the calculations will progressively tend to underestimate the critical concentration with decreasing uranium enrichment.

Experimental data are not available for assessment of the accuracy of the calculated limiting critical concentrations in solutions containing Th^{232}. The "finite concentration" cross sections for Th^{232} are expected to be as good as those for U^{238}, however, since the Th^{232} cross sections resulted from extensive studies in support of the ORNL Thorium Fuel Cycle Program.

The calculated limiting critical concentrations of pure U^{233} and Pu^{239} are 11.3 and 9.2 g/liter, respectively, as compared to experimentally determined best estimates of 11.2[3] and ~8.5[3] g/liter.

It is recommended that current process operations should approach no closer than within approximately 40% of the calculated concentrations. Design studies for processes that might operate several years from the present, by which time more experimental data should be available, could perhaps allow for maximum concentrations up to ~70% of the calculated critical concentrations.

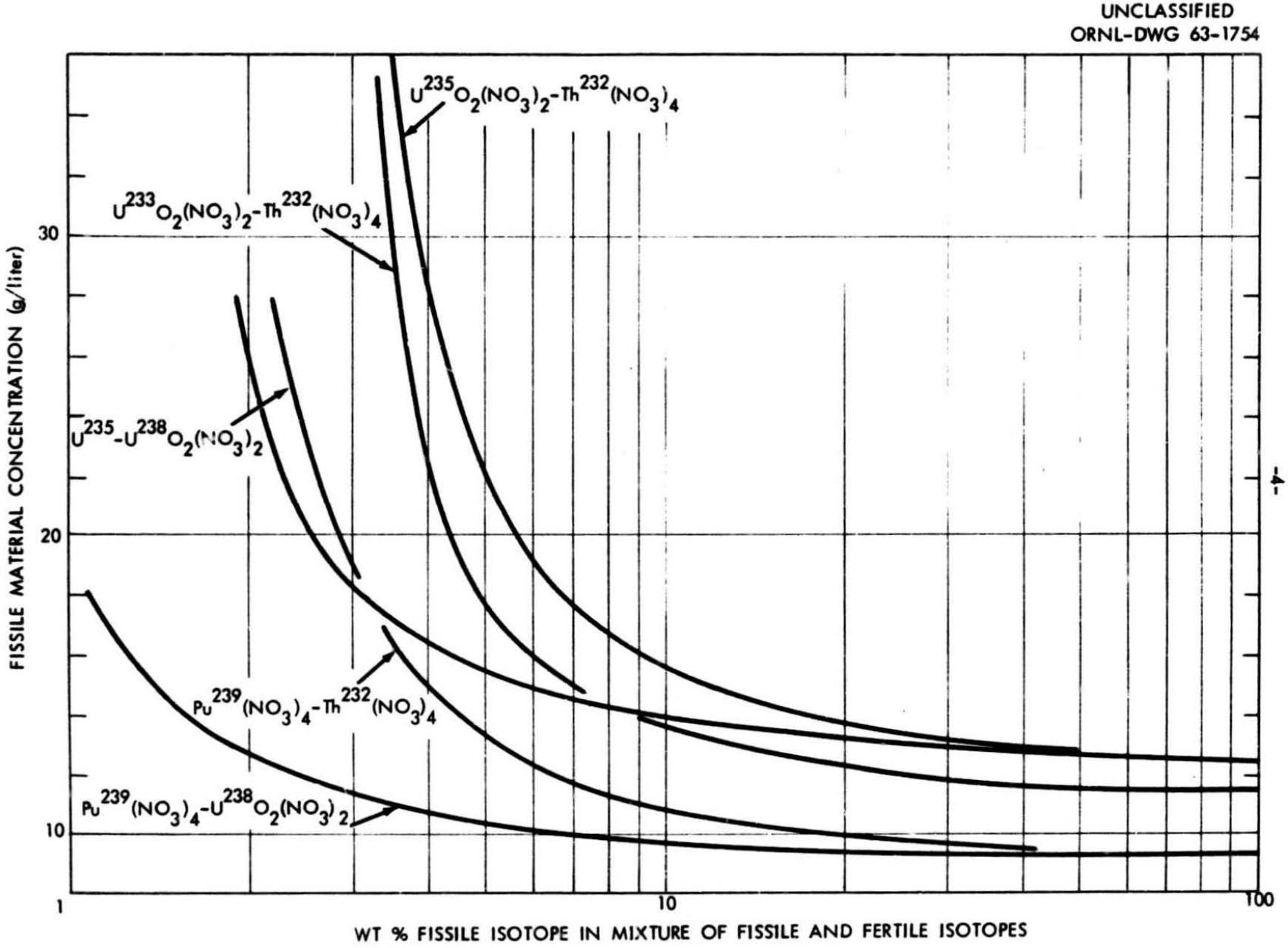

Fig. 1. Limiting Critical Concentrations of Aqueous Nitrate Solutions of Fissile and Fertile Isotopes.

Table 1. Summary of NDDRIC Calculations of the Limiting Critical Concentrations of U^{235} as U^{235}-$U^{238}O_2(NO_3)_2$ and $U^{235}O_2(NO_3)_2$-$Th^{232}(NO_3)_4$ in Natural Water

Enrichment Wt % U^{235} in U^{235}-U^{238} or Wt % U^{235} in U^{235}-Th^{232}	Calculated Critical Atomic Concentrations, atoms/barn-cm						Gms. U^{235} per liter	H/U^{235}
	H	O	N	U^{238}	Th^{232}	U^{235}		
2.00	3.905 E-2	4.586 E-2	6.575 E-3	3.284 E-3	-	6.660 E-5	25.98	586.3
2.619	5.102 E-2	4.055 E-2	3.760 E-3	1.985 E-3	-	4.972 E-5	19.39	1026
4.27	5.878 E-2	3.708 E-2	1.923 E-3	9.137 E-4	-	4.125 E-5	16.09	1425
8.93	6.352 E-2	3.498 E-2	8.040 E-4	3.622 E-4	-	3.597 E-5	14.03	1766
19.00	6.544 E-2	3.419 E-2	3.550 E-4	1.417 E-4	-	3.384 E-5	13.20	1934
29.49	6.597 E-2	3.392 E-2	2.230 E-4	7.795 E-5	-	3.302 E-5	12.88	1998
50.04	6.639 E-2	3.373 E-2	1.280 E-4	3.189 E-5	-	3.236 E-5	12.62	2052
70.36	6.657 E-2	3.366 E-2	8.70 E-5	1.334 E-5	-	3.207 E-5	12.51	2076
100.0	6.663 E-2	3.355 E-2	6.30 E-5	-	-	3.183 E-5	12.41	2093
3.569	4.690 E-2	5.330 E-2	9.910 E-3	-	2.430 E-3	6.880 E-5	34.64	528.1
5.254	5.970 E-2	4.200 E-2	4.050 E-3	-	9.870 E-4	5.403 E-5	21.06	1105
7.182	6.170 E-2	3.810 E-2	2.430 E-3	-	5.860 E-4	4.462 E-5	17.41	1360
9.956	6.350 E-2	3.640 E-2	1.530 E-3	-	3.620 E-4	3.966 E-5	15.47	1601
19.77	6.540 E-2	3.470 E-2	6.490 E-4	-	1.440 E-4	3.501 E-5	13.66	1868
49.03	6.640 E-2	3.390 E-2	1.990 E-4	-	3.320 E-4	3.260 E-5	12.72	2037

Table 2. Summary of MODRIC Calculations of the Limiting Critical Concentrations of U^{233}-Th^{232}, Pu^{239}-U^{238}, and Pu^{239}-Th^{232} as Nitrate Solutions in Natural Water

Wt % Fissile Isotope in Fissile plus Fertile Isotopes	Calculated Critical Atomic Concentrations (atoms/barn-cm)							Gms. Fissile Isotope Per Liter	H/U^{233} or H/Pu^{239}
	H	O	N	U^{238}	Th^{232}	U^{233}	Pu^{239}		
3.38	4.69 E-2	5.33 E-2	9.90 E-3		2.43 E-3	8.47 E-5		32.8	554
4.67	5.97 E-2	4.20 E-2	4.05 E-3		7.87 E-4	4.81 E-5		18.6	1240
6.43	6.17 E-2	3.81 E-2	2.43 E-3		5.86 E-4	4.01 E-5		15.5	1540
9.07	6.35 E-2	3.64 E-2	1.52 E-3		3.62 E-4	3.60 E-5		13.9	1770
18.2	6.54 E-2	3.47 E-2	6.44 E-4		1.44 E-4	3.19 E-5		12.3	2050
100 0	6.66 E-2	3.36 E-2	6.08 E-5		-	2.92 E-5		11.3	2280
1.24	3.90 E-2	4.59 E-2	6.64 E-3	3.22 E-3			4.04 E-5	16.0	966
1.78	5.10 E-2	4.06 E-2	3.79 E-3	1.83 E-3			3.30 E-5	13.1	1550
3.03	5.88 E-2	3.71 E-2	1.94 E-3	9.14 E-4			2.84 E-5	11.3	2070
6.51	6.35 E-2	3.50 E-2	8.26 E-4	3.62 E-4			2.51 E-5	9.97	2530
14.5	6.54 E-2	3.42 E-2	3.79 E-4	1.42 E-4			2.39 E-5	9.49	2740
23.4	6.60 E-2	3.39 E-2	2.50 E-4	7.80 E-5			2.37 E-5	9.41	2780
42.5	6.64 E-2	3.37 E-2	1.55 E-4	3.19 E-5			2.34 E-5	9.30	2830
100.0	6.63 E-2	3.36 E-2	9.04 E-5	-			2.32 E-5	9.22	2870
2.46	4.69 E-2	5.33 E-2	9.99 E-3		2.43 E-3		5.96 E-5	23.6	787
3.86	5.97 E-2	4.20 E-2	4.10 E-3		7.87 E-4		3.85 E-5	15.3	1550
5.34	6.17 E-2	3.81 E-2	2.47 E-3		5.86 E-4		3.21 E-5	12.7	1920
7.55	6.35 E-2	3.64 E-2	1.56 E-3		3.62 E-4		2.87 E-5	11.4	2210
15.39	6.54 E-2	3.47 E-2	6.77 E-4		1.44 E-4		2.54 E-5	10.1	2570
100.0	6.66 E-2	3.36 E-2	9.0 E-5		-		2.32 E-5	9.22	2870

Table 2. Summary of MODRIC Calculations of the Limiting Critical Concentrations of U^{233}-Th^{232}, Pu^{239}-U^{238}, and Pu^{239}-Th^{232} as Nitrate Solutions in Natural Water

Wt % Fissile Isotope in Fissile plus Fertile Isotopes	Calculated Critical Atomic Concentrations (atoms/barn-cm)							Gms. Fissile Isotope Per Liter	H/U^{233} or H/Pu^{239}
	H	O	N	U^{238}	Th^{232}	U^{233}	Pu^{239}		
3.38	4.69 E-2	5.33 E-2	9.90 E-3		2.43 E-3	8.47 E-5		22.8	554
4.67	5.97 E-2	4.20 E-2	4.05 E-3		7.87 E-4	4.81 E-5		18.6	12[illegible]
6.43	6.17 E-2	3.81 E-2	2.43 E-3		5.86 E-4	4.01 E-5		15.5	1540
9.07	6.35 E-2	3.64 E-2	1.52 E-3		3.62 E-4	3.60 E-5		13.9	1770
18.2	6.54 E-2	3.47 E-2	6.44 E-4		1.44 E-4	3.19 E-5		12.3	2050
100.0	6.66 E-2	3.36 E-2	6.08 E-5		-	2.92 E-5		11.3	2280
1.24	3.90 E-2	4.59 E-2	6.64 E-3	3.22 E-3			4.04 E-5	16.0	966
1.78	5.10 E-2	4.06 E-2	3.79 E-3	1.83 E-3			3.30 E-5	13.1	1550
3.03	5.88 E-2	3.71 E-2	1.94 E-3	9.14 E-4			2.84 E-5	11.3	2070
6.51	6.35 E-2	3.50 E-2	8.26 E-4	3.62 E-4			2.51 E-5	9.97	2530
14.5	6.54 E-2	3.42 E-2	3.79 E-4	1.42 E-4			2.39 E-5	9.49	2740
23.4	6.60 E-2	3.39 E-2	2.50 E-4	7.80 E-5			2.37 E-5	9.41	2780
42.5	6.64 E-2	3.37 E-2	1.55 E-4	3.19 E-5			2.34 E-5	9.30	2830
100.0	6.63 E-2	3.36 E-2	9.04 E-5	-			2.32 E-5	9.22	2870
2.46	4.69 E-2	5.33 E-2	9.99 E-3		2.43 E-3		5.96 E-5	23.6	787
3.86	5.97 E-2	4.20 E-2	4.10 E-3		7.87 E-4		3.85 E-5	15.3	1550
5.34	6.17 E-2	3.81 E-2	2.47 E-3		5.86 E-4		3.21 E-5	12.7	1920
7.55	6.35 E-2	3.64 E-2	1.56 E-3		3.62 E-4		2.87 E-5	11.4	2210
15.39	6.54 E-2	3.47 E-2	6.77 E-4		1.44 E-4		2.54 E-5	10.1	2570
100.0	6.66 E-2	3.36 E-2	9.0 E-5		-		2.32 E-5	9.22	2870

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