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CALCULATED LIMITING CRITICAL CONCENTRATIONS OF PLUTONIUM IN NATURAL URANIUM AND WATER

D. W. Magnuson*

ABSTRACT

Calculations have been performed to determine the limiting concentration of plutonium required for criticality when added to normal uranium. The bias of the method has been evaluated by comparing the calculated and experimental limiting ^{235}U enrichments for criticality of oxide-water mixtures and of nitrate-water mixtures.

INTRODUCTION

One of the important nuclear criticality safety parameters to be established for the processing of mixed oxide fuel is the amount of plutonium which can be added to a normal aqueous mixture with and without nitrate before criticality is achieved. Since there have been no experiments performed to determine these concentrations, calculations are being made to establish limits for these parameters. This memorandum summarizes a calculational effort which is one of several to be done for information to be included in the preparation of ANSI Standard ANS 8.12, Nuclear Criticality Control and Safety of Pu-U Fuel Mixtures Outside Reactors.

Calculational Method and Results

Previously, criticality calculations have been made using Hansen-Roach, 16 group cross sections in the ANISN transport code or in the KENO Monte Carlo code. Comparisons have been made to calculations using 123 group cross sections prepared from the ENDF/B Version II files without revealing any significant biases or trends in the differences. Therefore, the former method was used (HR 16 group cross sections in ANISN) to

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calculate k_∞ as a function of concentration for various values of uranium enrichment or plutonium fraction for UO_2-H_2O, $UO_2(NO_3)_2$-H_2O, UO_2-PuO_2-H_2O, and $UO_2(NO_3)$-$Pu(NO_3)_4$-H_2O mixtures. Near the maxima in the plots of the above data one calculation for each of the mixtures was repeated using the XSDRN transport code and 123 group cross sections. These calculated data are given in Tables 1 and 2 and are plotted in Figures 1 through 4.

The experimental and calculated critical uranium enrichments and the Pu critical concentrations in natural uranium are given in Table 3.

Table 3

LIMITING CALCULATED CRITICAL ENRICHMENT FOR URANIUM OR CRITICAL CONCENTRATION FOR PLUTONIUM

Mixture	Critical Enrichment (wt% ^{235}U)		Critical Concentration (wt% ^{239}Pu)	
	Experi-mental	Calcu-lated	Calcu-lated	Bias Corrected
UO_2-H_2O	1.034	1.010		
$UO_2(NO_3)_2$-H_2O	2.104	1.970		
$U(.71)O_2$-$^{239}PuO_2$-H_2O			0.159	0.183
$U(.71)O_2(NO_3)_2$-$^{239}Pu(NO_3)_4$-H_2O			0.678	0.712

The resultant biases in the calculations are 0.024 and 0.034 respectively for the oxide and nitrate mixtures.

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[1] U. I. Neeley and H. E. Handler, Measurement of the Multiplication Constant for Slightly Enriched UO_3-Water Mixtures and Minimum Enrichments for Criticality, HW-70310. Hanford Atomic Products Operation (1961).

[2] S. R. Bierman and G. M. Hess, Minimum ^{235}U Enrichment of Homogeneous Hydrogenous Uranyl Nitrate, Oak Ridge National Laboratory, ORNL-CDC-5 (June, 1968).

Table 1. Calculations of the Infinite Multiplication Factors for UO_2-H_2O and $UO_2(NO_3)_2$-H_2O Mixtures

Uranium Concentration (g/l)	Enrichment (wt% ^{235}U)	H:^{235}U Atom Ratio	Infinite Multiplication Factor - k_∞: ANISN 16 group	Infinite Multiplication Factor - k_∞: XSDRN 123 group
		UO_2-H_2O Mixtures		
2500	0.90	859	0.9342	
3000	0.90	665	0.9550	
3500	0.90	528	0.9602	
4000	0.90	424	0.9595	
2500	0.95	813	0.9561	
3000	0.95	630	0.9756	
3500	0.95	500	0.9804	
4000	0.95	402	0.9778	
2500	1.00	773	0.9765	
3000	1.00	599	0.9950	
3500	1.00	475	0.9986	0.9852
4000	1.00	382	0.9949	
		$UO_2(NO_3)_2$-H_2O Mixtures		
1100	1.85	733	0.9578	
1300	1.85	536	0.9742	
1500	1.85	392	0.9733	
1100	1.90	714	0.9691	
1300	1.90	522	0.9851	
1500	1.90	381	0.9835	
1100	1.95	695	0.9800	
1300	1.95	509	0.9961	0.9749
1500	1.95	371	0.9933	

Table 2. Calculations of the Infinite Multiplication Factors for H_2O and $U(nat)O_2(NO_3)_2$ - $^{239}PuO_2$-H_2O Mixtures

Natural Uranium Concentration (g/l)	Plutonium Concentration (g/l)	wt% Pu in (Pu+U)	Atom Ratio $H:^{235}U$	$H:^{239}Pu$	k_∞ 16 group ANISN	123 group XSDRN
		UO_2-PuO_2-H_2O Mixtures				
2500	3.5	0.14	1088	5610	0.9611	
3000	4.2	0.14	843	4348	0.9819	
3500	4.9	0.14	668	3447	0.9878	
4000	5.6	0.14	537	2771	0.9861	
2500	4.0	0.16	1088	4908	0.9757	
3000	4.8	0.16	843	3804	0.9958	
3500	5.6	0.16	668	3015	1.0009	
4000	6.4	0.16	537	2424	0.9984	
2500	4.5	0.18	1088	4363	0.9895	0.9846
3000	5.4	0.18	843	3381	1.0088	
3500	6.3	0.18	668	2680	1.0131	
4000	7.2	0.18	537	2154	1.0098	
		$UO_2(NO_3)_2$-$Pu(NO_3)_4$-H_2O Mixtures				
1200	6.6	0.55	1623	2133	0.9363	
1400	7.7	0.55	1186	1559	0.9486	
1600	8.8	0.55	858	1129	0.9480	
1200	7.2	0.60	1623	1954	0.9579	
1400	8.4	0.60	1186	1429	0.9688	0.9882
1600	9.6	0.60	858	1034	0.9673	
1200	7.8	0.65	1623	1805	0.9781	
1400	9.1	0.65	1186	1318	0.9882	
1600	10.4	0.65	858	955	0.9852	
1200	8.4	0.70	1623	1674	0.9969	
1400	9.8	0.70	1186	1223	1.0055	
1600	11.2	0.70	858	885	1.0007	

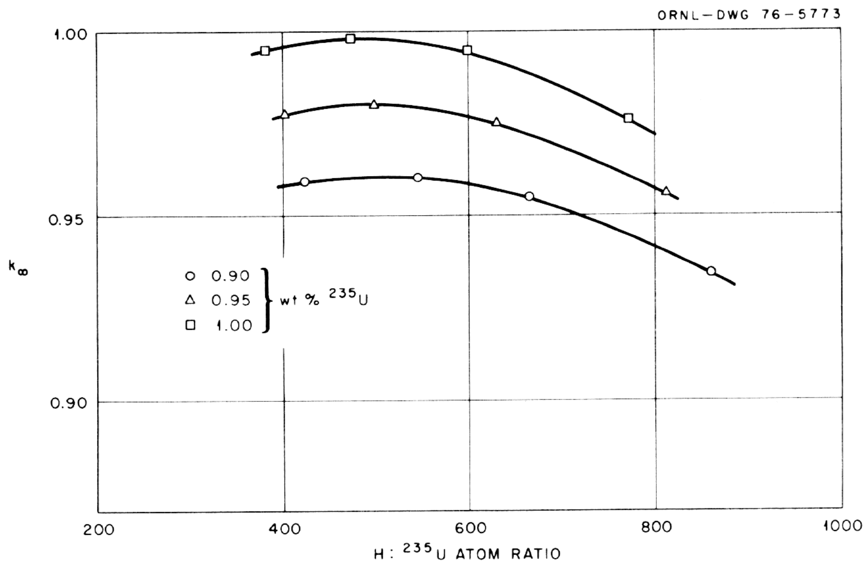

Fig. 1 k_∞ Calculated for UO_2-H_2O Mixtures

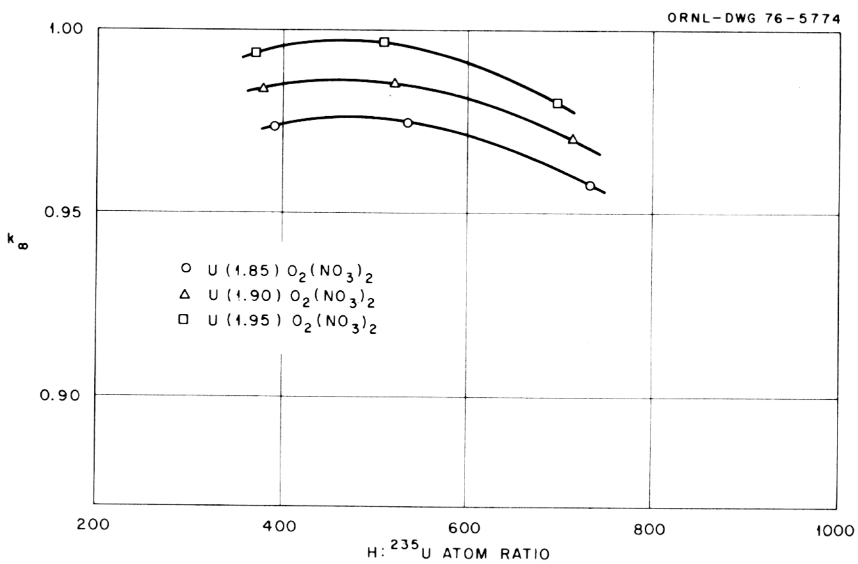

Fig. 2 k_∞ Calculated for $UO_2(NO_3)_2$-H_2O Mixtures

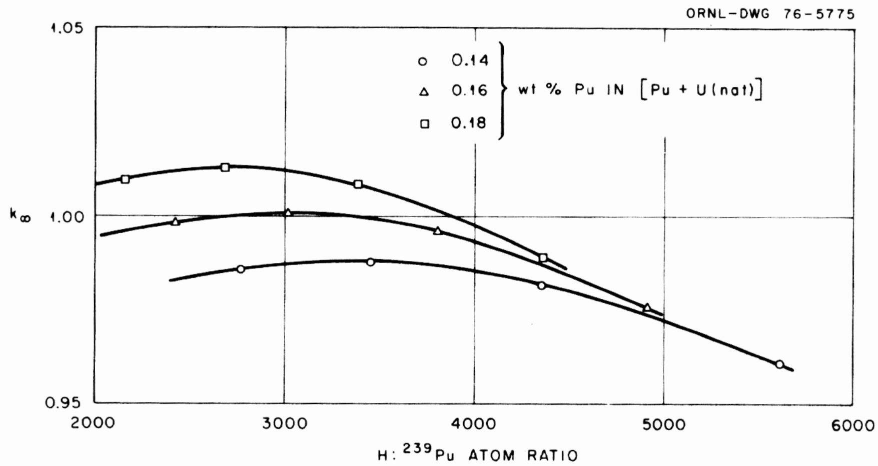

Fig. 3 k_∞ Calculated for $U(nat)O_2$-PuO_2-H_2O Mixtures

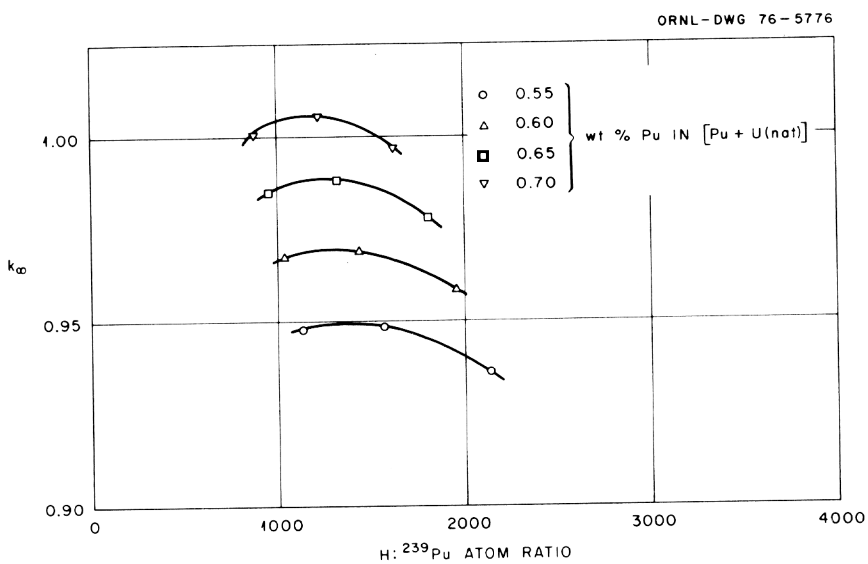

Fig. 4 k_∞ Calculated for $Pu(NO_3)_4$-$U(nat)O_2(NO_3)_2$-H_2O Mixtures

In comparison of oxide systems, it is seen that 0.30 wt% ^{235}U is equivalent to 0.159 wt% ^{239}Pu or a ratio of 1.887. It can be said that one atom of ^{239}Pu is equivalent to 1.887 atoms of ^{235}U. In the comparison of the nitrate systems, it is seen that one atom of Pu is equivalent to 1.873 atoms of ^{235}U. This value should be smaller because there is some neutron absorption by the two additional nitrogen atoms associated with each Pu atom; however, this effect is less than 0.4%, the ratio of the absorption cross sections of N and Pu. If the minimum critical concentrations are corrected for calculational biases, the values are 0.183 and 0.712 g of Pu per liter for oxide and nitrate, respectively.

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