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PARAMETRIC STUDIES OF THE REACTIVITY COEFFICIENTS FOR LARGE U²³³-Th-FUELED FAST REACTORS

by

R. S. Singh and H. H. Hummel

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ANL-6930

Reactor Technology

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AEC Research and
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Reactor Physics Division

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January 1966

*Formerly on loan from Indian Atomic Energy Establishment, Trombay,
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PARAMETRIC STUDIES OF THE REACTIVITY COEFFICIENTS FOR LARGE U^{233} -Th-FUELED REACTORS

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ABSTRACT

The feasibility of using U^{233} -Th as a fuel in a large fast breeder reactor has been studied from the point of view of the Doppler and sodium-void coefficients. The composition and temperature-dependent cross sections for thorium have been evaluated by using the latest resonance parameters available. The sodium-void coefficient has been studied in detail over a range of compositions and sizes of core for metal and oxide fuels, and the effect of the addition of U^{234} and fission products on the coefficient is also investigated. The reactivity coefficients due to the Doppler effect are calculated for metal- and oxide-fueled systems of interest. The results indicate that, even with present cross-section uncertainties, the reactivity changes due to both the sodium loss and the Doppler effect are encouraging for the development of fast breeder reactors with very large single cores if U^{233} and thorium are used as the fuel.

I. INTRODUCTION

The selection of a suitable fuel for a very large fast breeder reactor of the type to be used in connection with the desalination of water depends to a large extent on the nuclear properties of the fuel from the point of view of the temperature coefficients. It has been observed^{1,2} that the sodium-void coefficient in Pu^{239} - U^{238} -fueled fast reactors becomes positive as one goes to larger volumes of the core. The sodium-void coefficient arises mainly from the hardening of the neutron spectrum when the sodium atoms are expelled. The total coefficient is the sum of the three components of the effect: the spectral, the capture, and the leakage components.

The positive spectral component results from the decrease of the reactor capture-to-fission ratio with hardening of the spectrum. This decrease is because the Pu^{239} fission cross section varies more slowly with energy than do the capture cross sections of reactor materials. An additional positive effect is provided because U^{238} fissions are reduced by

inelastic scattering in sodium, so that U^{238} fissions increase as sodium is removed. The capture component, which is smaller than the spectral component, is due to the reduction in the capture of neutrons by sodium when it is removed from the core. These two positive components are compensated by the leakage component, which is negative and is a result of the increase in neutron leakage resulting from the increase in diffusion coefficient on loss of sodium atoms. In the case of large reactors, the leakage component is small and the enrichment is low; hence, the total sodium-void coefficient tends to be positive in large U^{238} -Pu-fueled fast reactors.

This problem is overcome to a large extent with U^{233} -Th as fuel because of the nuclear properties of these materials.³ The fission cross section of U^{233} varies more rapidly with energy than does that of Pu^{239} , and thorium has a much smaller fission cross section and a higher threshold energy for fission than U^{238} . As a consequence, the reactivity gain due to the spectral effect is much smaller with U^{233} -Th as fuel, compared to that with Pu^{239} - U^{238} as fuel. This important feature in U^{233} -Th systems encourages one to consider them as the fuel for very large fast breeder reactors. This study was made to investigate the details of the reactivity coefficients due to sodium loss and Doppler effect for such reactors as a function of core size and composition.

II. CROSS-SECTION PREPARATION

Most of the calculations for this study were done with a 22-group cross-section set, Set 215,⁴ prepared by using the first 13 groups of the YOM set⁵ with alterations as described below, and adding nine groups for the lower energies. Below Group 11 (15.25 keV), the composition and temperature dependence of heavy elements because of resonance self-shielding effects must be considered.

The effect of using cross sections from a newer set, Set 224,⁶ was also investigated.

A. Thorium

Resonance integral calculations were performed in the narrow resonance approximation. Temperature-dependent effective capture cross sections were obtained for the lower-energy groups below Group 11 covering both the unresolved and resolved resonance regions. The cross sections were evaluated for four temperatures: 300, 750, 1500, and 2500°K.

1. Set 215 Cross Sections

In the energy region up to 3 keV, resonance parameters for Th^{232} obtained at Columbia University⁷ were used to evaluate effective

resonance integrals. Since Γ_γ is not supplied for most of these resonances, a rather arbitrary procedure was adopted of using the same Γ_γ for a resonance as that for the next lowest in energy for which a value was supplied. This resulted in using values of Γ_γ mainly between 0.020 and 0.030 eV for resonances below 800 eV, and a value of 0.035 eV for resonances between 800 and 3000 eV. According to a private communication from H. Palevsky of Brookhaven National Laboratory, recent measurements of Γ_γ for the first eight strong levels in Th^{232} give an average value of 0.030 eV. An average value of Γ_γ of 0.0214 for the low-lying Th^{232} resonances was reported by Uttley and Jones.⁸

In the unresolved region from 3 to 25 keV (above which self-shielding was neglected), the following s-wave resonance parameters were used:

$$\langle \Gamma_n^0 \rangle = 1.27 \times 10^{-3} \text{ eV}, \quad D = 20.1 \text{ eV}, \quad \Gamma_\gamma = 0.0214, 0.040 \text{ eV}.$$

A Porter-Thomas distribution of neutron widths was assumed, ten discrete widths being used to describe the distribution. Values for σ_γ for both choices of Γ_γ are given in Table I for $\sigma_p = 40$ barns, a few values being given also for $\sigma_p = 60$ barns.

TABLE I. Contribution of s-wave to the Unresolved-region Capture Cross Section of Thorium (barns)

$\Gamma_n^0 = 1.27 \times 10^{-3} \text{ eV}, \quad D = 20.1 \text{ eV}$								
$\sigma_p = 40 \text{ barns}$								
$E_L, \text{ keV}$	$T = 300^\circ\text{K}$		$T = 750^\circ\text{K}$		$T = 1500^\circ\text{K}$		$T = 2500^\circ\text{K}$	
	$\Gamma_\gamma = 0.0214$	$\Gamma_\gamma = 0.040$	$\Gamma_\gamma = 0.0214$	$\Gamma_\gamma = 0.040$	$\Gamma_\gamma = 0.0214$	$\Gamma_\gamma = 0.040$	$\Gamma_\gamma = 0.0214$	$\Gamma_\gamma = 0.040$
15.0-25.0	0.1287	0.2075	0.1334	0.2157	0.1362	0.2206	0.1378	0.2236
9.1	0.1904	0.3006	0.2007	0.3181	0.2073	0.3292	0.2113	0.3362
4.0	0.2911	0.4459	0.3151	0.4851	0.3312	0.5117	0.3416	0.5292
2.8	0.4204	0.6224	0.4697	0.6992	0.5052	0.7555	0.5295	0.7945
$\sigma_p = 60 \text{ barns}$								
$\Gamma_\gamma = 0.040$								
$T = 1500^\circ\text{K}$			$T = 2500^\circ\text{K}$					
0.2254			0.2275					
0.3403			0.3456					
0.5399			0.5539					
0.8195			0.8528					

The p-wave contribution to the capture cross sections in the unresolved resonance region (Groups 12 to 15) was evaluated by choosing the strength function $\langle \Gamma_n^0 \rangle / D$ for p-wave resonances so that the calculated sum of s- and p-wave contributions gave 0.54 barn at 20 keV. This is at the upper limit of the data in Ref. 9 and agrees with the curve in Ref. 10. The value thus obtained for $\langle \Gamma_n^0 \rangle / D$ is 1×10^{-4} ; a value of 0.6×10^{-4} would

lower the calculated cross section at 20 keV by 0.1 barn. These calculations were done with the methods of Nicholson.¹¹ The p-wave contributions to σ_γ are given in Table II. These, together with the s-wave values for $\Gamma_\gamma = 0.040$ MeV and $\sigma_p = 40$ barns, were used in subsequent calculations. Cross sections for the resolved region below 2.8 keV are given in Table III. The choices made for Γ_γ in the energy range from 800 eV to 25 keV favor the higher values reported experimentally and tend to give more positive values of the sodium-void effect and higher values for the Doppler effect. The net change in the sodium-void effect for a lower Γ_γ might not be very much, however, after the necessary adjustment was made in the p-wave strength function. Use of a lower Γ_γ could substantially reduce the calculated Doppler effect, perhaps by as much as a factor of 1.5.

TABLE II. Contribution of p-wave to the Unresolved-region Capture Cross Section of Thorium (barns)

$$\begin{aligned} \langle \Gamma_n^0 \rangle / D &= 1.0 \times 10^{-4}, & D_{J=1/2} &= 20.1 \text{ eV}, \\ \Gamma_\gamma &= 0.030 \text{ eV}, & D_{J=3/2} &= 10.0 \text{ eV} \end{aligned}$$

E_L , keV	$\sigma_p = 40$ barns			
	T = 300°	T = 750°	T = 1500°	T = 2500°
15.0-25.0	0.3072	0.3102	0.3118	0.3122
9.1	0.3132	0.3167	0.3183	0.3193
4.0	0.2846	0.2880	0.2897	0.2907
2.8	0.2339	0.2367	0.2382	0.2391

TABLE III. Temperature-dependent Capture Cross Section of Thorium in Resolved Region (barns)

j	E_L , keV	$\sigma_p = 40$ barns				$\sigma_p = 60$ barns			
		300°K	750°K	1500°K	2500°K	300°K	750°K	1500°K	2500°K
16	2.0	0.995	1.144	1.253	1.328	1.138	1.277	1.373	1.436
17	1.4	1.348	1.577	1.759	1.894	1.607	1.837	2.013	2.138
18	1.0	1.07	1.254	1.395	1.498	1.251	1.431	1.565	1.660
19	0.55	1.088	1.323	1.528	1.692	1.334	1.594	1.812	1.978
20	0.30	1.03	1.288	1.54	1.758	1.310	1.628	1.929	1.914
21	0.10	1.28	1.60	1.93	2.28	1.65	2.08	2.52	2.90
22	0.03	0.723	0.829	0.958	1.087	0.919	1.076	1.255	1.430

Although a constant σ_p of 40 barns has been used in the calculations, higher values could well be appropriate in a given design, which would lead to a higher σ_γ and a more positive sodium-void effect.

In the energy region of importance for this effect, above ~ 1 keV, the change in σ_γ at 1500°K from $\sigma_p = 40$ to $\sigma_p = 60$, which represents a reasonable range for this parameter, is small compared to the change corresponding to a change in temperature from 300 to 1500°K, the result of which is shown in the calculations.

Fission and capture cross sections for thorium used in the calculations are summarized in Table IV. The values of σ_γ in Groups 9-11 in Set 215 have been lowered over the YOM values to correspond to the data of Ref. 9. Cross sections for groups 1-8 are identical with YOM values.

TABLE IV. Fission and Capture Cross Sections for Th^{232}
for $\sigma_p = 40$ barns

j	E_L , keV	ν	Set 215			Set 224			
			σ_f	$\sigma_\gamma(300^\circ\text{K})$	$\sigma_\gamma(1500^\circ\text{K})$	ν	σ_f	$\sigma_\gamma(300^\circ\text{K})$	$\sigma_\gamma(1500^\circ\text{K})$
1	3668	2.94	0.16	0.0197		2.74	0.170	0.0204	
2	2225	2.6	0.126	0.0394		2.37	0.128	0.0404	
3	1350	2.4	0.081	0.0744		2.22	0.095	0.0754	
4	825			0.12		2.15	0.034	0.128	
5	500			0.17				0.174	
6	300			0.18				0.18	
7	180			0.21				0.195	
8	110			0.27				0.242	
9	67			0.24				0.309	
10	41			0.32				0.423	
11	25			0.37				0.494	
12	15			0.515	0.532			0.518	0.533
13	9.1			0.614	0.647			0.606	0.636
14	4.0			0.730	0.801			0.711	0.776
15	2.8			0.855	0.993			0.675	0.804
16	2.0			0.995	1.253			0.744	0.949
17	1.4			1.348	1.759			0.915	1.208
18	1.0			1.070	1.395			0.974	1.258
19	0.55			1.088	1.528			0.956	1.334
20	0.30			1.030	1.540			0.960	1.771
21	0.10			0.895*	1.354*			1.641	2.413
22	0.03			0.723	0.958			1.033	1.289

*These numbers were found to be in error subsequent to the calculations. Correct values are 1.28 barns at 300°K and 1.93 barns at 1500°K. It is not expected that these errors would affect the results reported here significantly.

2. Set 224 Cross Sections

In this case, Γ_γ of Th^{232} was held constant at 0.030 eV. A p-wave strength function of 1.0×10^{-4} was used, which yielded a cross section of 0.53 barn at 20 keV at high dilution. These cross sections vary from the Set 215 cross sections mainly in that high values of σ_γ

have been assumed in Groups 9, 10, and 11. As will be seen in Section III-A, this produced a considerable difference in the calculated sodium-void effect.

B. Uranium-233

Only a limited amount of experimental information is available concerning capture cross sections and resonance parameters of U^{233} . Attempts were made to evaluate a reasonable value of $\langle \Gamma_f \rangle$, the average value of the fission width, by making some calculations of infinite-dilution cross sections based on the methods of Nicholson.¹⁰ The strength function $\langle \Gamma_n^0 \rangle / D$ was chosen to be 1×10^{-4} , and the fitting on $\langle \Gamma_f \rangle$ was made to obtain a capture-to-fission ratio, α , equal to 0.2 at 1 keV. The value of $\langle \Gamma_f \rangle$ so obtained was very high (~ 650 mV). Further investigations to obtain the shielded cross sections for U^{233} were not made, in view of the absence of reliable resonance parameters for this element, because of the problem of multilevel effects, and also because its contribution to the Doppler effect is probably negligible. Fission cross sections at low energies were taken from BNL-325.¹⁰

The Set 215 cross sections were prepared by using the same values as the YOM set⁵ for the first 13 groups, except for adjustment in the capture cross section to obtain agreement with the α measurements of Hopkins and Diven.¹² The values tabulated in the Russian cross-section sets¹³ were also consulted in preparing the low-energy capture cross sections for U^{233} . The α values assumed in the intermediate-energy region are rather high, which would tend to give a more positive sodium-void effect. The sodium-void effects calculated here are probably not very sensitive to the low-energy U^{233} α . A lower α would give a slightly higher thorium Doppler effect.

The Set 224 cross sections have a U^{233} fission cross section substantially lower than that for Set 215 because of a desire to be consistent with the lower fission cross section of U^{235} measured by White *et al.*¹⁴ The U^{233} fission cross section is normalized relative to that of U^{235} .

Lower values of α have also been used in the low-energy region, the assumed α values being the average of "high α " and "low α " values in Set 224, which were taken to represent extreme limits.

Fission and capture cross sections for U^{233} are listed in Table V.

C. Uranium-234

As the buildup of the heavier isotopes increases with the burnup of fuel, their influence on the reactivity coefficients has to be considered. Therefore, U^{234} and fission products were also included in our studies of the sodium-void coefficients. The cross sections for U^{234} given in Table VI

correspond to $\sigma_p = 1000$ and are based on the data given in Ref. 15 and on the Russian data.¹³ Alternate values of σ_γ suggested by Parker,¹⁶ also listed in Table VI, were also studied.

TABLE V. Fission and Capture Cross Sections for U^{233}

j	E_L , keV	Set 215			Set 224		
		ν	σ_f	σ_γ	ν	σ_f	σ_γ
1	3667	3.11	1.64	0.013	3.09	1.73	0.009
2	2225	2.85	1.84	0.023	2.83	1.83	0.018
3	1350	2.72	1.92	0.039	2.69	1.90	0.035
4	825	2.64	1.95	0.077	2.60	1.93	0.064
5	500	2.59	2.02	0.129	2.54	1.95	0.113
6	300	2.55	2.24	0.160	2.50	2.11	0.166
7	180	2.53	2.32	0.197	2.48	2.19	0.199
8	110	2.51	2.47	0.247	2.47	2.30	0.236
9	67	2.5	2.75	0.322	2.46	2.57	0.280
10	40.7	↓	3.13	0.416	2.45	3.11	0.333
11	25.0		3.63	0.54	2.45	3.77	0.39
12	15.0		4.26	0.68	↓	4.40	0.50
13	9.1		5.02	0.82		5.08	0.72
14	4.0		6.70	1.4		6.67	1.05
15	2.8		7.50	1.6		7.39	1.30
16	2.0		7.70	1.8		7.70	1.45
17	1.4		8.40	2.0		8.45	1.69
18	1.0		8.80	2.1		9.15	1.92
19	0.55		11.0	3.4		11.0	2.66
20	0.30		15.0	6.2		15.0	4.44
21	0.10		25.0	10.0		25.0	7.5
22	0.03		33.0	13.0		30.4	9.0

TABLE VI. Cross Sections for U^{234} and for Fission Products (barns)

j	E_L , keV	ν	σ_f	σ_γ	σ_γ (Parker)	σ_γ (F. P. Pairs)
1	3668	3.00	1.55	0.02	0.001	0.026
2	2225	2.77	1.52	0.04	0.003	0.039
3	1350	2.62	1.43	0.08	0.020	0.045
4	825	2.51	1.22	0.25	0.050	0.059
5	500	2.44	0.70	0.25	0.043	0.077
6	300	2.42	0.50	0.28	0.042	0.099
7	180	2.40	0.12	0.30	0.055	0.128
8	110		0.04	0.45	0.076	0.165
9	67		0.04	0.50	0.11	0.21
10	40.7		0.03	0.55	0.16	0.275
11	25.0		0.02	0.75	0.23	0.36
12	15.0		0.02	1.0	0.36	0.48
13	9.1		0.02	1.2	0.56	0.63
14	4.0		0.02	1.4	1.1	0.89
15	2.8			1.8	1.7	1.24
16	2.0			2.2	2.3	1.5
17	1.4			3.5	3.3	1.8
18	1.0			4.5	4.2	2.2
19	0.55			5.5		3.1
20	0.30			7.0		5.9
21	0.10			10.0		7.4
22	0.03			15.0		15.0

D. Fission Products

The capture cross sections of the fission products were evaluated for U^{233} yields by using the data provided by Garrison and Roos,¹⁷ with saturation effects neglected. These are given in Table VI. The elastic and inelastic scattering cross sections used are the same as those quoted for molybdenum in YOM.

Note

Fluxes and adjoint functions for Set 215 for the cases shown later in Figs. 6, 7, 10, and 11 were calculated with a σ_γ for sodium in Group 15 of 0.120 barn, as compared to 0.444 barn, the original Set 215 value, which was used in the other case. All capture components given correspond to the smaller value. This inconsistency should not affect other components of the sodium-void effect significantly.

III. SODIUM-VOID COEFFICIENT

To study the reactivity coefficients in large cores, three cases were considered corresponding to the volumes 6000, 8000, and 15,000 liters. Both metal and oxide fuels with volume fractions of 70% sodium, 15% stainless steel, and 15% fuel were studied. To compare the effect of the coolant volume fraction on the sodium-void coefficient, one case with volume fractions 50% sodium, 20% stainless steel, and 30% fuel was studied. The effect of adding U^{234} and fission products on the sodium coefficient was also investigated. The specifications of the enrichments and the compositions for the various cores and blankets are given in Table VII.

The method employed to obtain the effective group cross sections for transport and elastic transfer corresponding to the nonfissile and non-fertile materials in the different cases studied was to use the ELMOE¹⁸ program. This program solves a fundamental mode problem by using hundreds of fine groups and takes into account the details of the elastic-scattering resonances of the light elements, the data on light-element, angular-dependent scattering cross sections being provided by a library of Legendre polynomial coefficients. The effective "coarse-group" cross sections are obtained for the different cases with full, half, and no sodium-volume fractions in them, at enrichment ratios in the range of interest.

The ELMOE code also provides the 22 coarse-group fluxes, which are normalized such that

$$\sum_j (\nu \Sigma_f)_j \phi_j = 1.$$

TABLE VII. Specifications of the Systems Studied

Metal fuel density:	18.57 g/cc (for U^{233}) 11.58 g/cc (for Th^{232})
Oxide fuel density:	8.4 g/cc
Core volume fractions:*	70% sodium 15% stainless steel 15% fuel (Th and U^{233})
Blanket volume fractions:	20% thorium 15% stainless steel 65% sodium
Blanket thickness:	54 cm in all cases
Reflector volume fractions:	40% sodium 60% stainless steel
Reflector thickness:	30 cm (extrapolated) in all cases

In the cases in which U^{234} is present,** $\frac{\text{atoms of } U^{234}}{\text{atoms of } U^{233}} = 0.4$.

Fission-product pairs when present correspond to 5 a/o of fuel.

	$\frac{\text{Atoms/cc} \times 10^{-24}}{\text{at Full Density}}$
U^{233}	0.048
Th^{232}	0.030 (as metal) 0.0192 (as oxide)
Stainless steel	0.0847
Sodium	0.022

*Except in cases where 50 v/o Na, 20 v/o SS and 30 v/o fuel are considered.

**Based on Ref. 20.

Another code evaluates the adjoint fluxes for these cases, which are normalized in accordance with

$$\sum_j \beta_j \phi_j^+ = 1,$$

where β_j is the fraction of fission spectrum in group j . With this normalization of the fluxes, the spectral and capture components of the reactivity coefficients are calculated by perturbation theory by means of the cross sections derived from ELMOE. The spectral component is the sum of the elastic and inelastic removal effects due to the hardening of the spectrum when sodium is removed from the core. The capture component due to the change in capture with the loss of sodium is usually small. These three components are evaluated by perturbation calculations for the loss of sodium from full to half and from half to nil. The values for full to half are obtained with the full-sodium and half-sodium fluxes, and an average of the two is taken. The values for half to nil are similarly obtained with half-sodium and no-sodium fluxes, and the average of the two values is again taken. These coefficients for all the cases under study are tabulated in Table VIII.

TABLE VIII. Spectral and Capture Components of Sodium-void Effect
Sodium 70 v/o unless otherwise indicated

Fuel Type	Th ²³² + U ²³⁴ U ²³³	% Δk, Full to Half (Full Sodium Flux)				% Δk, Full to Half (Half Sodium Flux)				% Δk, Half to Nil (No Sodium Flux)				Spectral and Capture	
		Elastic Removal	Capture Comp.	Inelastic Removal	Spectral plus Capture	Elastic Removal	Capture Comp.	Inelastic Removal	Spectral plus Capture	Elastic Removal	Capture Comp.	Inelastic Removal	Spectral plus Capture	Full-to-Half Average	Half-to-Nil Average
Metal No U ²³⁴ 300°K	6.0 6.8 8.0 12.5 (50% Na)	-2.09 -1.64 -1.17 1.05	0.13 0.15 0.17 0.07	-0.24 -0.15 -0.03 -0.24	-2.20 -1.64 -1.03 1.36	-2.49 -2.06 -1.37 0.95	0.10 0.12 0.14 0.06	-0.34 -0.23 -0.07 -0.27	-2.73 -2.17 -1.30 1.28	-4.05 (0.03) -3.59 (0.05) -2.77 (0.07) 0.83 (0.07)	0.08 0.10 0.12 0.06	-0.76 -0.62 -0.39 0.27	-4.73 -4.11 -3.04 1.16	-2.47 -1.91 -1.17 1.32	-3.73 -3.14 -2.17 1.22
Metal U ²³⁴ 300°K	7.0 8.9 8.0 (F.P. + U ²³⁴)	-0.83 0 0.12	0.13 0.17 0.18	0.04 0.23 0.18	-0.66 0.40 0.48	-1.12 0.10 0.09	0.11 0.15 0.15	0.03 0.28 0.23	-0.98 0.53 0.47	-2.07 (0.05) -0.54 (0.09) -0.56 (0.18)	0.09 0.12 0.12	-0.09 0.27 0.26	-2.07 -0.15 -0.18	-0.82 0.47 0.48	-1.53 0.19 +0.15
Oxide No U ²³⁴ 300°K	4.4 5.5 6.5 8.0 (F.P. + U ²³⁴)	-3.16 -3.02 -3.26 -0.78	0.23 0.28 0.34 0.42	-0.05 -0.03 -0.02 0.16	-2.98 -2.77 -2.94 -0.20	-3.66 -3.40 -3.21 -0.39	0.22 0.25 0.32 0.42	-0.06 -0.04 -0.03 0.22	-3.50 -3.19 -2.92 0.25	-5.37 (0.20) -4.70 (0.22) -3.86 (0.20) +1.29 (0.77)	0.27 0.35 0.34 0.66	-0.11 -0.08 -0.06 0.26	-5.21 -3.813 -3.58 2.21	-3.24 2.98 -2.93 0.03	-4.36 -3.51 -3.25 0.26
Metal F.P. + U ²³⁴ 1500°K	8.0	1.05	0.17	0.22	1.44	0.85	0.14	0.25	1.24	-0.26 (0.23)	0.12	0.26	0.12	1.34	0.68
Metal F.P. + U ²³⁴ 1500°K 50 v/o Na	11.8	1.46	0.05	0.33	1.84	1.39	0.05	0.36	1.80					1.82	
Oxide F.P. + U ²³⁴ 1500°K	8.0	0.47	0.39	0.26	1.12	1.15	0.39	0.31	1.85	3.71 (2.62)	0.60	0.33	4.64	1.49	
Metal F.P. + U ²³⁴ 300°K (Set 224) 1500°K	8.0 8.0	0.13 1.09	0.35 0.313	0.31 0.36	0.79 1.78	0.52 1.08	0.30 0.29	0.41 0.45	1.23 1.82	0.77 (0.34) 0.99 (0.41)	0.25 0.28	0.61 0.49	1.63 1.76	1.01 1.27	1.43 1.25
Oxide F.P. + U ²³⁴ 300°K (Set 224) 1500°K	8.0 8.0	-0.31 1.13	0.65 0.62	0.29 0.42	0.63 2.17	-0.09 1.74	0.68 0.64	0.41 0.53	1.00 2.91	2.77 (2.57) 5.14 (3.52)	0.95 0.89	0.61 0.72	4.33 6.75	0.82 2.54	

The total sodium-void coefficient is evaluated by a space-dependent calculation using the REX code. The critical enrichment ratios are evaluated for the reactors of given core volumes and blanket specifications;

the k_{eff} for the same systems with only half sodium and with no sodium present in them is evaluated. The composition of the blanket and the reflector used in the problems is given in Table VII. The leakage component is obtained by subtracting the spectral-plus-capture effect from the total reactivity coefficient. These results are given in Table IX.

TABLE IX. Components of Sodium-void Effect in Spherical Cores

Fuel Type	% Na	Volume, liters	$\frac{Th^{232} + U^{234}}{U^{233}}$, Approx	% Δk , Full to Half			% Δk , Half to Nil		
				Total	Spectral and Capture	Leakage	Total	Spectral and Capture	Leakage
Metal, No U^{234} , $T = 300^\circ K$	70	6,000 8,000 15,000	6.6 7.1 8.9	-3.7	-2.1 -1.7 -0.6	-3.1	-5.9	-3.2 -2.9 -1.4	-4.5
Metal + U^{234} , $T = 300^\circ K$	70	8,000 15,000	7.0 9.0	-2.4	-0.8 +0.5	-2.9	-4.4	+0.2	-4.6
Oxide, No U^{234} , $T = 300^\circ K$	70	6,000 15,000	5.5 7.5	-6.0	-6.8 -2.9	-3.8 -3.1	-8.3	-3.5	-4.8
Metal + U^{234} + F.P., $T = 300^\circ K$	70		8.0		+0.5			+0.2	
Oxide + U^{234} + F.P., $T = 300^\circ K$	70		8.0		0.0			+0.3	
Metal, No U^{234} , $T = 300^\circ K$	50	15,000	11.8	-0.6	+1.1	-1.7	-0.9	+1.3	-2.2
Metal (F.P. + U^{234}), $T = 1500^\circ K$	70	15,000	8.1	-1.3	+1.5	-2.8	-4.5	+0.8	-5.3
Metal (F.P. + U^{234}), $T = 1500^\circ K$	50	15,000	11.7	+0.7	+1.8	-1.1			

A. Results with Set 215 and Set 224

It is noted in Table VIII that the spectral-plus-capture components for metal- as well as oxide-fueled cores (70 v/o sodium) are negative when neither U^{234} nor fission products are added to them. Also, the spectral and capture effects increase with decreasing enrichment (Fig. 1), as has been observed earlier for Pu- U^{238} systems.¹⁹ This is partly because the ratio of sodium to fuel is higher at lower enrichment for fixed volume fractions of fuel and sodium, as was assumed here. For the capture component it is also because there is more low-energy flux under these conditions, and for the spectral component it is because the adjoint flux varies more sharply with energy at lower enrichment. The more rapid variation of the adjoint function with energy is mainly because leakage, which depresses

Fig. 1. Spectral and Capture δk in Th^{232} - and U^{233} -fueled Cores for Full to Half Sodium, 300°K

the high-energy adjoint function, is less at lower enrichment. The spectral-plus-capture components for the core with 50 v/o sodium are seen to be positive. This behavior is due to the low enrichment in this case, with consequent low leakage.

These effects can be observed in Figs. 2, 3, and 4. In Fig. 2, the energy dependence of η , the fission neutrons emitted per neutron absorbed in the core, is given for various enrichments. The variation of η with energy is observed not to be very marked and not to change much with enrichment. In Fig. 3, the effect of leakage has been incorporated for the metal-fueled cases of Fig. 2 to yield what is denoted as $k_{\text{eff}}(E)$, the fission neutrons emitted per neutron being absorbed or leaking at energy E . In Fig. 4, the adjoint functions are given for the same cases as those in Fig. 3, the adjoint function

differing from $k_{\text{eff}}(E)$ in that the effect of energy transfers has been added, greatly smoothing out the effect of variations in $k_{\text{eff}}(E)$. The spectral components for the 12.5 Th/U^{233} case at 50 v/o sodium are seen in Table VIII to be positive. This is associated with the rise in adjoint function with energy above 10 keV for this case (Fig. 4).

Fig. 2. Fission Neutrons Produced per Neutron Absorbed, η , versus E , 300°K , No Fission Products or U^{234} , Full Sodium

Fig. 3. Multiplication Factor, k_{eff} , versus E , Metal Fuel, 300°K, No Fission Products or U^{234} , Full Sodium

Fig. 4. Adjoint Function, Metal Fuel, 300°K, Full Sodium, No Fission Products or U^{234}

The addition of U^{234} equal to 0.4 of the U^{233} based on Ref. 20 and fission-product pairs (5 a/o of fuel) to metal as well as oxide changes the sign of the spectral component at 300°K. This can again be explained as due to the change in adjoint flux variation (Fig. 5) with the addition of these materials, which have an assumed capture-cross-section variation more rapid than the fission cross section of U^{233} , causing a more rapid variation of η with energy.

Fig. 5. Adjoint Function, Metal Fuel, 300°K, 70 v/o Sodium, $8.0(\text{Th}^{232} + \text{U}^{234})/\text{U}^{233}$, Full Sodium, Showing Effect of Removing Fission Product and U^{234} Capture

Raising the assumed Th^{232} temperature from 300 to 1500°K increases the low-energy Th^{232} capture cross section, thereby causing a more rapid variation of the adjoint function (Fig. 6) and therefore a more positive sodium-void effect. A temperature of 1500°K is too high for metal fuel and may be high for oxide, but one can interpolate for the desired temperature. There is a large change in adjoint-function variation and in sodium-void effect from the 300°K case with no U^{234} or fission products present, to the 1500°K case with these materials present. This

Fig. 6. Adjoint Function, Metal Fuel, 300 and 1500°K, 70 v/o Sodium, $8.0(\text{Th}^{232} + \text{U}^{234})/\text{U}^{233}$, Full Sodium, Fission Products and U^{234} Present

points to the desirability of being able to make critical assembly measurements of sodium-void effects at high fuel temperatures, although it is possible to obtain information on the temperature variation of the sodium-void effect from Doppler-effect measurements at varying sodium content. It is obviously also important that the actual isotopic composition of the fuel, including fission products, be mocked up as faithfully as possible.

In some cases the spectral component tends to be more negative for oxide systems than for a metal-fueled system. This is caused by the fact that the adjoint function varies more slowly because of the stronger moderation, the low-energy region below 1 keV with higher η and small leakage being more strongly coupled to the region above 1 keV. This corresponds physically to the fact that, as moderation increases, a neutron slowing down above 1 keV has a greater probability of surviving to lower energies.

The data in Table VIII indicate that strong nonlinearities occur in the elastic-removal component of the sodium-void effect as a function of fraction of sodium removed for some of the cases with 70 v/o sodium. These nonlinearities are due mainly to the change in energy variation of the adjoint function with sodium content (Fig. 7), and to a much smaller extent to changes in the energy spectrum of the flux (Figs. 8 and 9). An exception to this statement is the strong positive effect at no sodium caused in some cases by the 2.85-keV resonance in sodium. This effect is indicated in parentheses in Table VIII following the elastic-removal values for no sodium. The quantity given is the contribution to the elastic-removal cross section for Group 15, which contains the resonance. The large values are caused by the fact that the no-sodium flux does not contain

Fig. 7. Adjoint Function, Metal Fuel, 1500°K, 70 v/o Sodium, $8.0(\text{Th}^{232} + \text{U}^{234})/\text{U}^{233}$, Fission Products and U^{234} Present, for Full, Half, and No Sodium

Fig. 8. Flux for Metal Fuel, 1500°K, 70 v/o Sodium, $8.0(\text{Th}^{232} + \text{U}^{234})/\text{U}^{233}$, Fission Products and U^{234} Present, for Full, Half, and No Sodium

Fig. 9. Flux for Oxide Fuel, 1500°K, 70 v/o Sodium, $8.0(\text{Th}^{232} + \text{U}^{234})/\text{U}^{233}$, Fission Products and U^{234} Present, for Full, Half, and No Sodium

a flux dip associated with the resonance. As sodium is introduced and the dip appears, the contribution from Group 15 will be quickly reduced and should not contribute much to the integrated sodium-removal effect. The effect is much larger for certain oxide systems because of the larger low-energy flux. It is smaller in other oxide cases because the adjoint flux variation from Group 15 to Group 16 happens to be very small in such cases. When the Group 15 contribution is very large, no average half- to no-sodium value is presented since more detailed information is evidently needed. Changes in sodium cross sections with fraction of sodium removed have played a negligible role in these nonlinearities. Table X gives effective elastic-removal cross sections for sodium as determined from ELMOE calculations for full to half sodium and half to no sodium. These two sets of values, which are seen to be virtually identical, are obtained by subtracting in one case the group macroscopic elastic-removal cross section for half sodium present from that at full sodium, and in the other that for no sodium from that for half sodium, dividing the difference in either case by the number of atoms of sodium removed.

TABLE X. Effective Elastic-removal Cross Sections of Sodium

j	Metal, Th/U = 8, No U ²³⁴ and F.P., T = 300°K		Oxide, Th/U = 8, U ²³⁴ + F.P. Present, T = 300°K	
	Full to Half	Half to Nil	Full to Half	Half to Nil
2	0.296	0.291	0.318	0.320
3	0.262	0.261	0.268	0.268
4	0.506	0.527	0.491	0.518
5	0.838	0.875	0.826	0.861
6	0.742	0.728	0.737	0.736
7	0.710	0.701	0.711	0.734
8	0.745	0.759	0.685	0.699
9	0.804	0.790	0.770	0.777
10	0.789	0.806	0.803	0.846
11	0.763	0.782	0.760	0.784
12	0.731	0.747	0.728	0.740
13	0.850	0.798	0.867	0.864
14	0.970	1.006	0.990	1.047
15	15.19	16.66	15.25	17.13
16	2.595	3.130	2.627	3.207
17	0.999	1.001	0.997	0.999
18	0.820	0.818	0.818	0.818
19	0.403	0.383	0.422	0.418
20	0.398	0.362	0.430	0.424
21	0.124	0.080	0.141	0.112

The nonlinearity at 50 v/o sodium is less than at 70 v/o because of the smaller ratio of sodium to fuel, giving less change in flux and adjoint spectrum with removal of sodium.

For the calculations with Set 215 at $8.0(\text{Th} + \text{U}^{234})/\text{U}^{233}$ with U^{234} and fission products, for metal fuel the elastic-removal component of the sodium-void effect becomes much more negative as sodium is removed,

but for oxide fuel this component becomes more positive. This surprising behavior appears to have the following explanation: The comparatively low values for σ_γ of Th^{232} assumed for Groups 9, 10, and 11 lead to high values of the adjoint function in these groups for the metal case with no sodium (see Fig. 7). This leads to large negative contributions to the sodium-void effect in this region, which is one of relatively large flux. For the cases with sodium, the moderation present is strong enough to smooth out the adjoint function. For oxide, this effect is less because of the moderating effect of the oxygen (Figs. 10 and 11). In both cases, the void effect tends to become more positive with removal of sodium, for the following reason: When the moderating effect of sodium is removed, the low-energy fissions, with higher η , have less effect on the slope of the adjoint function at higher energies, above a few keV, where the bulk of the sodium-void effect occurs.

Fig. 10

Adjoint Function for Metal and Oxide Fuel with Set 215 Cross Sections and Metal Fuel with Set 224 Cross Sections, 1500°K, 70 v/o Sodium, 8.0($\text{Th}^{232} + \text{U}^{234}$)/ U^{233} , Fission Products and U^{234} Present, for Full Sodium

Fig. 11
Adjoint Function for Metal and Oxide Fuel with Set 215 Cross Sections and Metal Fuel with Set 224 Cross Sections, 1500°K, 70 v/o Sodium, 8.0($\text{Th}^{232} + \text{U}^{234}$)/ U^{233} , Fission Products and U^{234} Present, for No Sodium

In the Set 224 cross sections, because of the different assumption made about the energy variation of σ_γ of Th^{232} , the disparity between the behavior of metal and oxide cases with removal of sodium is less.

Because of the uncertainty in cross sections, particularly of the σ_γ of Th^{232} , the nonlinearities reported here should be regarded only as illustrative of what effects may occur. They do suggest that a careful study of the spectral component of the sodium-void effect in various thorium-fueled systems with differing amounts of moderation may indicate how smoothly varying the Th^{232} capture cross section is. The same statement applies to U^{238} -fueled systems, for which the energy variation of the adjoint function is also rather uncertain. It is significant that rather small changes (of the order of several percent) in the adjoint function from one group to another in the region of maximum flux, produce changes of the order of a percent in the elastic-removal component of the sodium-void effect.

The leakage component of the sodium-void effect, given in Table IX, decreases with increasing size, and is expected to be about the same for a given core size, and fuel type, and core volume fractions. For this it can be inferred that the reactivity effect for total removal of sodium from the core is negative for all the cases listed in Table IX, except for the one for 15,000 liters core volume at 1500°K with 50 v/o sodium and with fission products and U^{234} present. Nonlinearities are observed in the leakage component as well as in the spectral component when sodium content varies from full to half and from half to nil.

The results obtained here for the sodium-void effect are encouraging enough that very large fast breeders for desalination based on U^{233} -Th fuel seem very likely to be feasible from this standpoint, even when the effect of cross-section uncertainties is considered. The spatial dependence of the various components of the sodium-void effect has not been obtained here but would have to be considered in detail safety studies of a particular system.

B. Effect of Cross-section Variation

The effects of certain variations in cross sections from those of Set 215 have been studied in the 8:1 metal system with fission products and U^{234} at full sodium. The effect of substituting the Parker σ_γ values¹⁶ of U^{234} for the Set 215 values did not make any appreciable difference in the sodium-void effect and affected the adjoint function only slightly. The use of the Set 224 inelastic-transfer matrices instead of those for Set 215 did not affect the results appreciably. Changing the low-energy α values of U^{233} to those of Set 224 made no appreciable difference. The effect of the use of the complete Set 224 cross sections has already been discussed in Section A above.

IV. DOPPLER EFFECT

The importance of the Doppler coefficient, which may be the only reliable, prompt-acting, negative-reactivity feedback available in large fast reactors, has been recognized by many people. The Doppler effect is due mostly to neutrons with energies below about 10 keV; therefore its effect is more pronounced in the systems with softer spectra. An idea of the magnitude of the effect²¹ can be had by calculating the slowing-down density 'q' of neutrons at 9.1 keV. In the U^{233} -Th systems we have studied, this is found to be large even with metal fuel, predicting a substantial Doppler coefficient in them. The spectrum is soft in the U^{233} -Th metal-fueled cores because of the lower density of the fuel and higher volume fraction (70%) of sodium used in our calculations. The ELMOE program¹⁸ was used for our flux calculations because this more accurately treats the moderation and scattering by light elements contributing to the lower-energy flux in fast reactors. Of course, this involves a fundamental mode calculation and therefore does not take into account the spatial dependence of the flux or the temperature distribution in the core.

The calculations have been performed for the three core volumes with different enrichments for both metal-fueled and oxide-fueled cores, which are the same systems considered for the calculations of sodium-void coefficient. The problems are first run with the room-temperature (300°K) cross sections of thorium to obtain a critical value of the buckling B^2 , and they are rerun with the cross sections at higher temperature using the same buckling B^2 , to obtain k_{eff} . The change in reactivity Δk in going from one temperature to another is obtained therefrom. All the cases were also studied with no sodium present in the core.

The results of these calculations are given in Table XI. It can be seen that the values of $\Delta k/\Delta T$ obtained for the metal-fueled systems are large, comparable in magnitude to those that can be obtained in Pu^{239} - U^{238} oxide-fueled systems. The large Doppler coefficient in metal systems available with the sodium present in the core is reduced by a factor of around 15 when the sodium is removed from the core, and thus becomes almost negligible. This can be attributed to the sharp decrease in the low-energy spectrum in the core when the sodium is expelled, which is more pronounced in a metal-fueled system with large volume fraction of sodium present. This is a disadvantage in these metal systems from the safety point of view, but the disadvantage can be overcome by adding a small amount of moderating material like BeO, as suggested by Greebler.²² The coefficients for the oxide-fueled core are quite large, and they increase with decreasing enrichment. The reduction in the Doppler coefficient when the sodium atoms are expelled is only by a factor of five, as has been observed earlier⁴ in cores with 70 v/o sodium. This smaller reduction is due to the softer spectrum present in the oxide-fueled core.

TABLE XI. Reactivity Change % Δk due to Doppler Effect, by Direct k Calculations

Fuel Type	Th/U ²³³	Sodium	300-750°K		750-1500°K		1500-2500°K		q
			-% Δk	$-\frac{\Delta k}{\Delta T} \times 10^5$	-% Δk	$-\frac{\Delta k}{\Delta T} \times 10^5$	-% Δk	$-\frac{\Delta k}{\Delta T} \times 10^5$	
Metal	6.0	Yes	0.55	1.22	0.38	0.51	0.25	0.25	0.093
		No	0.04	0.09	0.02	0.03	0.01	0.01	0.003
	6.8	Yes	0.70	1.56	0.48	0.78	0.32	0.32	0.114
		No	0.04	0.09	0.03	0.04	0.02	0.02	0.005
	8.0	Yes	0.92	2.04	0.63	0.84	0.42	0.42	0.143
		No	0.07	0.16	0.04	0.05	0.02	0.02	0.007
Oxide	4.4	Yes	0.98	2.18	0.73	0.98	0.52	0.52	0.201
		No	0.24	0.53	0.16	0.21	0.10	0.10	0.049
	5.5	Yes	1.33	2.96	0.98	1.31	0.71	0.71	0.246
		No	0.36	0.80	0.23	0.31	0.15	0.15	0.069
	6.5	Yes	1.72	3.82	1.29	1.72	0.92	0.92	0.293
		No	0.52	1.16	0.38	0.51	0.15	0.15	0.093

Table XII lists Doppler effects obtained by perturbation calculation for the cases of Table XI plus some additional ones. The difference between the results with 300 and 1500°K fluxes indicates that perturbation theory is in error for this large a change, so that perhaps an average of

TABLE XII. Reactivity Change % Δk due to Doppler Effect for Temperature Changes from 300 to 1500°K by Perturbation Calculation

Fuel Type	Th ²³² + U ²³⁴ U ²³³	Flux Used					
		Full Sodium		Half Sodium		No Sodium	
		300°K	1500°K	300°K	1500°K	300°K	1500°K
Metal	6.0	1.03		0.40		0.049	
	6.8	1.32		0.54		0.064	
	8.0	1.76		0.75		0.095	
	12.5 (50% Na)	0.93		0.44		0.13	
Metal + U ²³⁴	7.0	1.06		0.42		0.052	
	8.9	1.73		0.74		0.096	
	8.0 (F.P. + U ²³⁴)	1.45	1.18	0.63	0.51	0.085	0.071
Oxide	4.4	1.87		1.17		0.40	
	5.5	2.55		1.69		0.62	
	6.5	3.30		2.29		0.89	
	8.0 (F.P. + U ²³⁴)	3.60	3.26	2.57	2.24	1.11	0.95
Metal, (F.P. + U ²³⁴) 50 v/o Na	11.8		0.47		0.23		

the two should be used. The results with 300°K flux are therefore all too high by the order of 10%. The Doppler effects in Table XII appear to be consistent with the temperature variation of the sodium-void effect shown in Table VIII, although comparison is difficult because of the strong nonlinearities present.

ACKNOWLEDGMENT

We wish to thank Dr. D. Okrent for his suggestions and keen interest in this work. Extensive assistance from Mr. A. L. Rago, Miss Lois Meyer, and Miss Joann Zeman, in carrying out machine calculations, and from Mrs. Stella Dean with hand calculations is gratefully acknowledged.

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