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A BURNUP SURVEY OF Pu-Th-BeO FUELS AND COMMENTS ON SELECTION OF FUEL FOR PEBBLE BED REACTORS

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November 1966

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ABSTRACT

The results of a burnup survey for a BeO pebble bed reactor system studied by the Australian Atomic Energy Commission are reported. The survey was based on an equilibrium model and carried out by means of the GYMEA complex of codes. A once-through fuel cycle was assumed with plutonium and thorium as the fissile and fertile materials. The selection of a fuel composition is discussed taking into consideration reactivity lifetime, initial power density, and temperature coefficients of reactivity. With a 200 MWe pebble bed reactor design, values of F.I.F.A. (fissions per initial fissile atom) of 1.2 to 1.4 appear to be achievable at an average bulk power density of 11 W/cm^3.

CONTENTS

1. INTRODUCTION

In the reference design of a BeO pebble bed reactor system studied by the Australian Atomic Energy Commission (Ebeling and Hayes 1966) plutonium and thorium in the form of oxides were specified as the fissile and fertile materials respectively. In this report a study of the burnup of a once-through fuel system is described and the results are given of surveys carried out by means of the GYMEA complex of codes (Pollard and Robinson 1966) in September 1965. Fuel recycling is treated in a separate report (Bicevskis 1966).

2. PROCEDURE ADOPTED

The survey was based on the equilibrium spectrum model (Bicevskis and Hesse 1966) as incorporated in the GYMEA code.

Within the framework of GYMEA, the nuclear data were provided by NDXB cross section and NDSC scattering libraries which contain 55 nuclides (34 fission products) and 120 energy groups.

As the input to GYMEA, one specified, besides the initial fuel composition, the required average power density and F.I.F.A. (fissions per initial fissile atom). The code then produced the corresponding values of average material buckling.

The bulk of the investigations were carried out using an average power density of 11 W/cm^3 as specified for the upflow version of the reference design. A few representative runs were repeated at 7 and 15 W/cm^3 for optimization study of the core size.

3. SPECIFICATION OF COMPOSITIONS

The voidage of a random packed pebble bed is 0.4. With the concentrations of Pu and Th considered, the atomic density of BeO is practically constant and an approximate value of 4×10^{22} atoms cm^{-3} was used throughout.

The two independent variables are the atomic densities of Pu and Th which were used for interpreting the results.

For convenience the atomic densities have been expressed in units of 10^{19} atoms cm^{-3} and a short-hand notation adopted. As an example, a composition with a plutonium atomic density of 3×10^{19} atoms cm^{-3} and a thorium atomic density of 20×10^{19} atoms cm^{-3} is described as 3/20.

Use of atomic ratios of fissile:fertile:moderator material to specify the fuel composition has the following disadvantages for our survey:

(i) It is difficult to convert from fissile and fertile material ratios to atomic densities; the latter must be used in the burnup calculations.

(ii) "Fertile" material is defined as both Th232 and Pu240. These nuclides have vastly different properties (the cross section of Pu240 is larger by a factor of approx. 70).

(iii) For our system, the fuel is dispersed in all the moderator, and core size is fixed by the permissible power density. Thus by changing the "moderator ratio" one really changes the fissile investment.

For our study, the following Pu isotopic composition has been specified (nominal 3000 MWd/tonne exposure):

Pu239	-	78 per cent.
Pu240	-	17 per cent.
Pu241	-	5 per cent.

This gives a fissile material concentration in Pu of 83 per cent. With the above values, one obtains the following approximate conversion formulae:

$$\text{Moderator Ratio} = \frac{4000}{0.83 \times N_{Pu}} \approx \frac{4800}{N_{Pu}} ,$$

$$\text{Fertile Ratio} = \frac{N_{Th} + 0.17\ N_{Pu}}{0.83\ N_{Pu}} ,$$

where:

N_{Pu} = atomic density of Pu in 10^{19} atoms cm^{-3}, and

N_{Th} = atomic density of Th in 10^{19} atoms cm^{-3}.

4. DISCUSSION OF RESULTS

The main results of the survey are given in Figures 1 to 6 and Tables 1 to 7. The results are largely self-explanatory but the following qualifications and comments are provided:

4.1 Effect of Plutonium and Thorium Concentrations

If the concentration of Pu is kept constant and the Th content is varied (Figures 1-3), one obtains the pattern observed previously by E.W. Hesse (unpublished) that with increasing Th investment the buckling curves tend to flatten out with the average value of the buckling falling. For large Th concentrations the buckling curves are flat, but the systems are sub-critical.

If the Th concentrations are kept constant but the Pu content is varied (Figures 4-6) one obtains a slightly increased average buckling as the Pu concentration is increased up to the point where the Pu concentration starts to approach the Th concentration. At this stage, for small values of F.I.F.A. the average buckling is still large but it drops sharply for large values of F.I.F.A. as the Th is depleted.

As a generalization, the thorium concentrations have much larger effect on average buckling than the plutonium concentrations within the range of compositions investigated.

4.2 Achievable Burnups and Selection of Compositions from Reactivity Considerations

Assuming a required average material buckling of 2.1×10^{-4} cm^{-2} for the reference core design (E.W. Hesse 1966b) it follows from Figures 1 to 6 and Table 1 that the burnups obtainable, as governed by reactivity lifetime, would be approximately 1.4 F.I.F.A. The best plutonium concentrations range from 3 (in our units) upwards, with only a slight increase in buckling with increasing Pu concentration. In terms of moderator ratios, this would mean ratios from 1:1600 downward. The best thorium concentration appears to be 20, irrespective of the fissile concentration.

Based on reactivity considerations, one of the best choices appears to be a composition 3/20 or in the "old" notation, 1:8:1600. The "reference" composition 1:16.5:1650 has a thorium concentration of 40 and, as shown in Table 1, it would lead to a subcritical reactor, even for small values of F.I.F.A.

The composition 3/20 was used as a basis for investigating neutron balances, cross sections, etc.

Some check runs were made with the NDXC cross section library which has 120 energy groups and 100 nuclides, including 78 fission products. It also has an explicit treatment for the Sm150 chain. With the NDXC library a value for F.I.F.A. of approx. 1.2 for the composition 3/20 was obtained.

The equivalent system buckling of 2.1×10^{-4} cm^{-2} can only be considered an approximation as some of the design details have not been finalized. It appears reasonable to assume for preliminary estimates, that values of F.I.F.A. within the range 1.2 to 1.4 are achievable with the specified reference design.

4.3 Approximate Relations Between Reactivity, F.I.F.A., and Power Density

The following approximate relations were derived for the composition 3/20

at a value for F.I.F.A. of 1.4 and power density of 11 W/cm^3.

A reactivity of 1 per cent. is equivalent to a change in buckling of 0.33×10^{-4} cm^{-2} and 0.04 in F.I.F.A.

The results of Table 1 were obtained for the power density of 11 W/cm^3 specified in the reference design. For the optimization study, some typical compositions were also investigated at power densities of 7 and 15 W/cm^3 and the results are given in Table 2. On the average, for an increase of 4 W/cm^3 in the power density, there is a decrease of approx. 0.4×10^{-4} cm^{-2} in buckling and a decrease of approx. 0.05 in F.I.F.A.

The above approximations should not be used for large departures from the reference values.

4.4 Neutron Balances

Some typical neutron balances are given in Table 3. The selection is based on perturbing one factor at a time from the reference case of 3/20 composition at a F.I.F.A. of 1.4 and power density of 11 W/cm^3. A careful perusal of this table will reveal many interesting features of the survey.

It is to be noted that for 3/20 composition, the absorption in fission products takes up 7.0, 10.2, and 12.4 per cent. respectively, as the burnup is increased from a value for F.I.F.A. of 1.0 to 1.4 and 1.8.

4.5 Initial Power Density

The initial, average, and exit power densities are also given in Table 3.

As pointed out by E.W. Hesse previously, low thorium concentrations lead to high initial power densities. As an example, for the same Pu atomic density of 3×10^{19} atoms cm^{-3} and a value for F.I.F.A. of 1.4, an increase in the Th atomi density from 20 to 30×10^{19} atoms cm^{-3} reduces the initial power density from 40.86 to 25.91 W/cm^3.

The power output of the BeO pebbles is limited by the permissible thermal stresses. A space dependent fuel management study is required for a realistic evaluation of this problem (Hesse 1966a).

4.6 Microscopic Absorption Cross Sections and Average Concentrations

Table 4 gives the microscopic cross sections and eta-values as a function of F.I.F.A. and Table 5 gives the average and ex. compositions. In general, cross sections increase with F.I.F.A. (with a few exceptions of very slight decreases).

It is interesting to note from Table 5 that the average concentration of U233 is higher for a burnup of 1.4 than for either 1.0 or 1.8. On the other hand, the exit concentration decreases steadily within the range 1.0 to 1.8 F.I.F.A. This is understandable in the light of the increasing microscopic absorption cross section of U233 (Table 4) and the increasing flux level.

The "Recovery Factor" also listed in Table 5 is defined as the ratio of exit to initial fissile material concentration. Two values have been provided; one excludes all Pa233 and the other includes all Pa233 as fissile material (100 per cent. decay). The difference is not large for the two extremes and any practical case would lie between these values.

Table 4 shows the superior η-value of U233 and Pu241 compared with Pu239 and U235.

4.7 Temperature Coefficients of Reactivity

The temperature coefficients of reactivity are given in Table 6 for the composition 3/20 as a function of F.I.F.A. The values were obtained as a difference from GYMEA criticality calculations at 900°K and 300°K.

For a more realistic assessment one should carry out space dependent criticality calculations at various core temperature covering both the normal operating range as well as accident conditions (failure of cooling system, etc.). Experiments with hot critical assemblies are required to ensure reliability of the predictions.

Table 7 displays the reactivity investments between 300°K and 900°K for a range of compositions.

In all cases, the temperature coefficient of reactivity becomes less negative with an increase in burnup and with a decrease in thorium concentration.

For the composition 3/20 the temperature coefficient of reactivity is slightly positive at a value for F.I.F.A. of 1.4. If this is unacceptable for plant control, one can achieve a slightly negative temperature coefficient of reactivity by adopting a composition 4/30 which also gives a value for F.I.F.A. of 1.4.

5. SUMMARY

1. For the reference BeO pebble bed reactor design with Pu as fissile and Th as fertile material the survey indicates that, from reactivity lifetime considerations, values of F.I.F.A. of 1.2 to 1.4 should be obtainable.

2. One of the best compositions is 3/20 (Pu atomic density 3×10^{19} atoms cm^{-3} and Th atomic density 20×10^{19} atoms cm^{-3}) or 1:8:1600 (fissile:fertile: moderator).

3. The temperature coefficients of reactivity become less negative with increasing F.I.F.A. and with decreasing thorium concentration.

4. The composition 3/20 has a slightly positive temperature coefficient of reactivity at a value for F.I.F.A. of 1.4. A negative value can be achieved by adopting a composition 4/30 (1:9:1200).

5. The initial power density also reduces with increasing thorium concentration.

6. ACKNOWLEDGEMENTS

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The authors of the GYMEA code, Messrs. Pollard and Robinson, adapted the code as required for the survey.

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TABLE 1 - AVERAGE BUCKLING V. F.I.F.A.
GYMEA SURVEY, SEPTEMBER 1965

Power Density = 11W/cm^3

Atomic Densities (10^{19} atoms cm^{-3})		Average Buckling (10^{-4} cm^{-2}) for various F.I.F.A.				
Pu	Th	1.0	1.2	1.4	1.6	1.8
1	10	3.49	1.54	-0.07	-1.07	-1.69
	20			-0.33	-0.98	-1.46
2	10	5.71	3.46	0.76	-0.68	-1.54
	20	3.37	2.64	1.50	0.35	-0.46
	30	1.24	0.98	0.52	-0.07	-0.63
	40	-0.75	-0.77	-0.90	-1.13	-1.41
	50	-2.60	-2.48	-2.41	-2.42	-2.49
3	10	6.37	4.13	0.83	-0.82	-1.79
	16	5.00	4.03	1.95	0.29	-0.72
	20	4.12	3.48	2.12	0.63	-0.34
	30	2.04	1.89	1.41	0.61	-0.16
	40	0.17	0.22	0.11	-0.20	-0.64
	50	-1.57	-1.39	-1.29	-1.34	-1.51
4	10	6.69	4.37	0.66	-1.09	-2.13
	16	5.30	4.40	2.02	0.20	-0.93
	20	4.48	3.88	2.34	0.66	-0.45
	30	2.51	2.38	1.86	0.89	-0.04
	40	0.72	0.79	0.66	0.29	-0.28
	50	-0.93	-0.74	-0.65	-0.74	-0.98
5	10	6.75	4.44	0.42	-1.44	-2.49
	16	5.52	4.59	1.95	-0.03	-1.24
	20	4.70	4.11	2.42	0.50	-0.68
	30	2.83	2.68	2.12	1.02	-0.07
	40	1.13		1.03		-0.11
	50	-0.44		-0.20		-0.63
10	20	5.31	4.401	2.168	-0.62	-2.13

AVERAGE BUCKLING V. POWER DENSITY FOR VARIOUS FUEL COMPOSITIONS

TABLE 2

Atomic Densities (10^{19} atoms cm^{-3})		F.I.F.A.	Average Buckling (10^{-4} cm^{-2}) for Various Power Densities (W/cm^3)		
Pu	Th		7	11	15
2	30	1.4	1.03	0.52	0.06
3	10	1.0	6.65	6.37	6.18
	10	1.4	1.18	0.83	0.48
	20	1.0	4.47	4.12	3.83
	20	1.4	2.54	2.12	1.73
	30	1.0	2.46	2.04	1.72
	30	1.4	1.86	1.41	1.01
	40	1.0	0.59	0.17	-0.21
4	30	1.4	0.58	0.11	-0.30
5	30	1.4	2.30	1.86	1.49
		1.4	2.51	2.12	1.78

TABLE 3

NEUTRON BALANCES (PERCENTAGE ABSORPTION) AND POWER DENSITIES (Atomic densities in units of 10^{19} atoms cm^{-3})

Variable		F.I.F.A.			Th			Pu				Average Power Density (W/cm^3)		
Atomic density	Pu	3	3	3	3	3	3	2	3	4	5	3	3	3
	Th	20	20	20	10	20	30	20	20	20	20	20	20	20
F.I.F.A.		1.0	1.4	1.8	1.4	1.4	1.4	1.4	1.4	1.4	1.4	1.4	1.4	1.4
Be9		-4.50	-4.03	-3.28	-2.51	-4.03	-4.41	-3.62	-4.03	-4.30	-4.43	-4.03	-4.03	-4.02
O16		1.03	1.25	1.61	1.98	1.25	1.08	1.45	1.25	1.13	1.06	1.24	1.25	1.26
Li6		1.24	2.19	2.84	2.89	2.19	1.77	2.16	2.19	2.21	2.21	2.16	2.19	2.22
He3		0.04	0.13	0.28	0.22	0.13	0.09	0.09	0.13	0.18	0.23	0.20	0.13	0.10
Th232		17.65	19.37	22.07	13.93	19.37	24.28	21.44	19.37	18.19	17.30	19.28	19.37	19.45
Pa233		0.62	0.90	1.41	1.08	0.90	0.91	1.17	0.90	0.74	0.65	0.58	0.90	1.22
U 233		6.51	11.25	16.33	10.18	11.25	12.16	11.29	11.25	11.23	11.19	11.47	11.25	11.04
U 234		0.30	0.73	1.57	1.03	0.73	0.68	0.65	0.73	0.80	0.87	0.62	0.73	0.83
U 235		0.06	0.29	1.05	0.71	0.29	0.20	0.25	0.29	0.33	0.37	0.25	0.29	0.35
Pu239		34.27	25.12	18.65	24.39	25.12	24.51	24.80	25.12	25.19	25.17	25.42	25.12	24.85
Pu240		19.01	14.84	11.0	14.34	14.84	14.41	14.50	14.84	14.96	15.00	15.01	14.84	14.69
Pu241		15.98	16.22	12.2	15.90	16.22	14.87	15.70	16.22	16.40	16.47	16.37	16.22	16.08
Pu242		0.77	1.54	1.86	2.23	1.54	1.13	1.11	1.54	1.88	2.15	1.52	1.54	1.55
Fission products		7.04	10.19	12.38	13.61	10.19	8.30	9.01	10.19	11.04	11.73	9.92	10.19	10.38
Total flux, 10^{14} $sec^{-1}cm^{-2}$		2.28	2.63	3.25	3.68	2.63	2.41	2.02	2.63	2.46	2.37	1.66	2.63	3.64
Av. B^2, 10^{-4} cm^{-2}		4.12	2.12	-0.336	0.83	2.12	1.41	1.49	2.12	2.34	2.42	2.55	2.12	1.73
Power Density, W/cm^3														
Initial		20.19	40.86	76.46	106.1	40.86	25.91	39.26	40.86	40.45	40.83	25.63	40.45	57.35
Average		11.04	10.99	10.94	10.92	10.99	10.94	10.95	10.99	10.90	10.98	7.07	10.90	14.98
Exit		6.52	4.63	5.72	3.52	4.63	6.12	5.14	4.63	4.33	4.17	2.98	4.33	6.30

TABLE 4

MICROSCOPIC ABSORPTION CROSS SECTIONS V. F.I.F.A.

Pu = 3×10^{19} atoms cm^{-3}, Th = 20×10^{19} atoms cm^{-3}, Power density 11 W/cm^3.

	Microscopic Absorption Cross Section (Barns) for Various F.I.F.A.		
	1.0	1.4	1.8
Be9	-0.00434	-0.00345	-0.00239
O16	0.000964	0.00104	0.00115
Cl2	0.00106	0.00114	0.00123
Li6	59.83	99.6	149.1
Th232	3.503	3.484	3.466
Pa233	48.97	55.98	63.19
U233	69.64	92.21	120.0
U234	34.09	35.38	37.0
U235	45.30	72.23	106.0
Pu239	174.8	305.5	459.7
Pu240	210.3	253.2	278.1
Pu241	147.3	248.7	372.7
Pu242	24.33	22.97	21.43
Cdll3	2723	5480	8899
Xel35	140600	301000	506600
Pml47	111.6	111.7	111.7
Sml49	3687	7668	12740
	η Values		
U233	2.160	2.185	2.205
U235	1.838	1.927	1.975
Pu239	1.801	1.813	1.822
Pu241	2.206	2.193	2.186

TABLE 5

ATOMIC DENSITIES OF MAIN NUCLIDES V. F.I.F.A.

Initial Composition Pu = 3×10^{19} atoms cm^{-3}, Th = 20×10^{19} atoms cm^{-3}, Power Density 11W/cm^3.

	Atomic Densities* $\left(10^{24} \text{ atoms } cm^{-3} \text{ or } \frac{1}{\text{barn.cm}}\right)$ for Various F.I.F.A.					
	1.0		1.4		1.8	
	Average	Exit	Average	Exit	Average	Exit
Li6	7.971E-7	1.369E-6	7.570E-7	1.079E-6	5.553E-7	6.567E-7
Th232	1.942E-4	1.886E-4	1.908E-4	1.820E-4	1.858E-4	1.723E-4
Pa233	4.862E-7	4.948E-7	5.508E-7	5.431E-7	6.523E-7	6.211E-7
U233	3.602E-6	6.286E-6	4.193E-6	6.155E-6	3.972E-6	4.763E-6
U234	3.365E-7	7.819E-7	7.040E-7	1.482E-6	1.241E-6	2.222E-6
U235	4.819E-8	1.547E-7	1.405E-7	3.978E-7	2.886E-7	6.511E-7
Pu239	7.551E-6	1.244E-6	2.829E-6	5.997E-9	1.184E-6	6.142E-14
Pu240	3.508E-6	1.209E-6	2.000E-6	4.458E-8	1.154E-6	1.744E-10
Pu241	4.178E-6	3.458E-6	2.244E-6	2.184E-7	9.550E-7	4.966E-10
Pu242	1.215E-6	2.387E-6	2.294E-6	2.852E-6	2.529E-6	2.040E-6
Cdll3	8.640E-10	6.175E-10	3.728E-10	4.734E-11	1.505E-10	1.158E-11
Xel35	4.289E-10	2.391E-10	2.230E-10	9.023E-11	1.185E-10	6.050E-11
Pml47	2.157E-7	2.404E-7	2.467E-7	1.818E-7	2.341E-7	1.847E-7
Sml49	4.802E-9	2.542E-9	1.914E-9	6.063E-10	8.739E-10	3.575E-10
Recovery Factor (excluding all Pa233)	-	0.448	-	0.231	-	0.217
Recovery Factor (including all Pa233)	-	0.467	-	0.294	-	0.242

* In this table the letter E stands for the base 10

For example, 7.971E-7 $\equiv 7.971 \times 10^{-7}$.

TABLE 6

TEMPERATURE COEFFICIENTS OF REACTIVITY V. F.I.F.A.

Pu = $\beta \times 10^{19}$ atoms cm^{-3}, Th = 20×10^{19} atoms cm^{-3}, Power density 11W/cm^3

F.I.F.A.	Multiplication Factor, k		$k_{900}-k_{300}$	$\frac{k_{900}-k_{300}}{900-300}$
	300°K	900°K		
1.0	1.1340	1.1096	-0.0244	-4.1×10^{-5}
1.4	1.0457	1.0614	+0.0157	$+2.6 \times 10^{-5}$
1.8	0.9452	0.9886	+0.0434	$+7.2 \times 10^{-5}$

TABLE 7

TEMPERATURE REACTIVITY INVESTMENTS

Difference in k (%) for core temperatures of 900°K and 300°K

Atomic Density, 10^{19} atoms cm^{-3}		F.I.F.A.			
Pu	Th	1.0	1.2	1.4	1.8
2	20	+0.17		+5.19	
3	16	-1.97		+3.47	
	20	-2.44	-0.86	+1.57	+4.34
	30	-3.41		-1.24	
4	16	-3.09		+2.40	
	20	-3.51		+0.31	
	30	-4.37		-2.48	
5	16	-3.67		+1.62	
	20			-0.55	

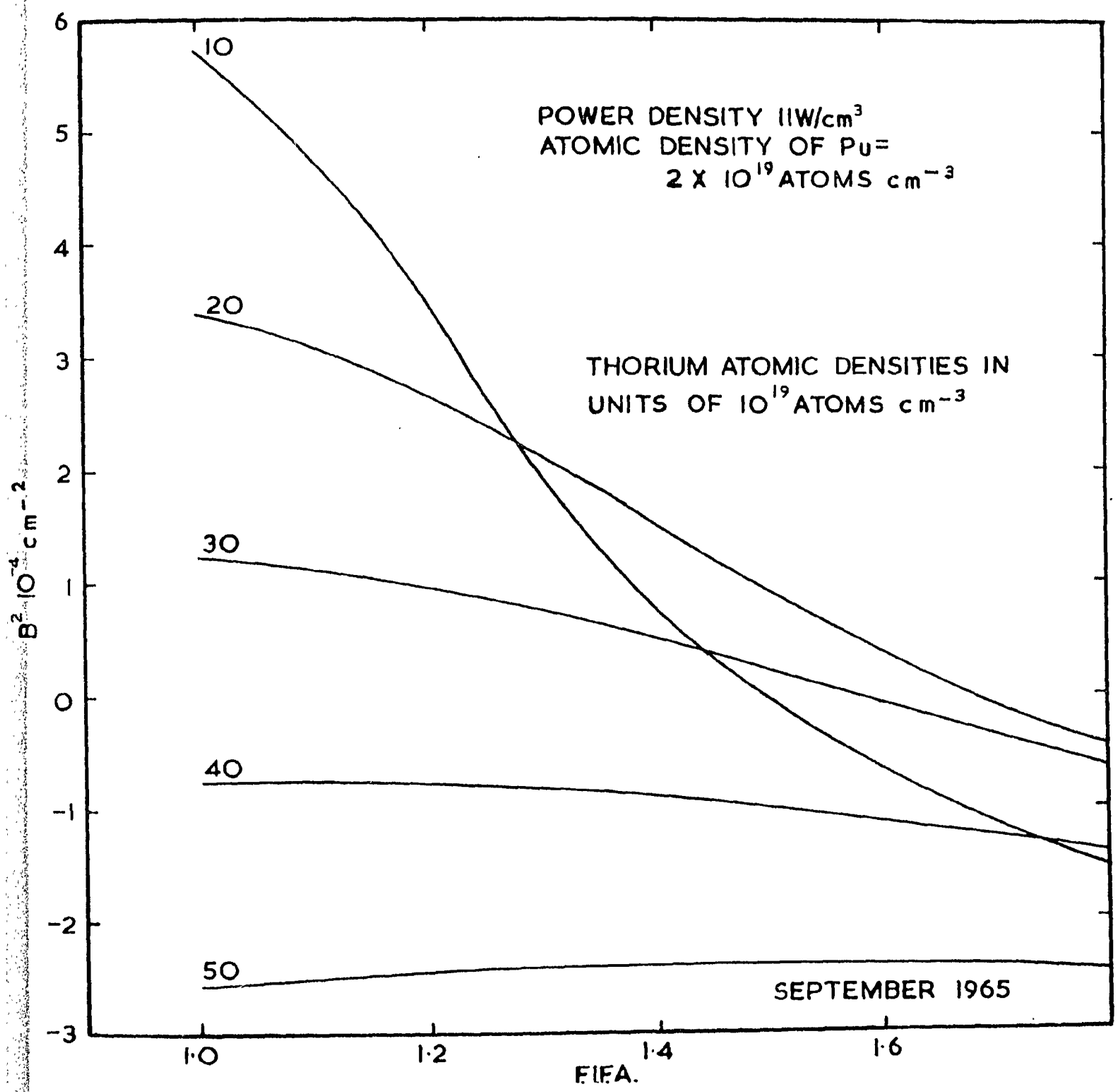

FIGURE 1. AVERAGE MATERIAL BUCKLING v. F.I.F.A.

P849/1

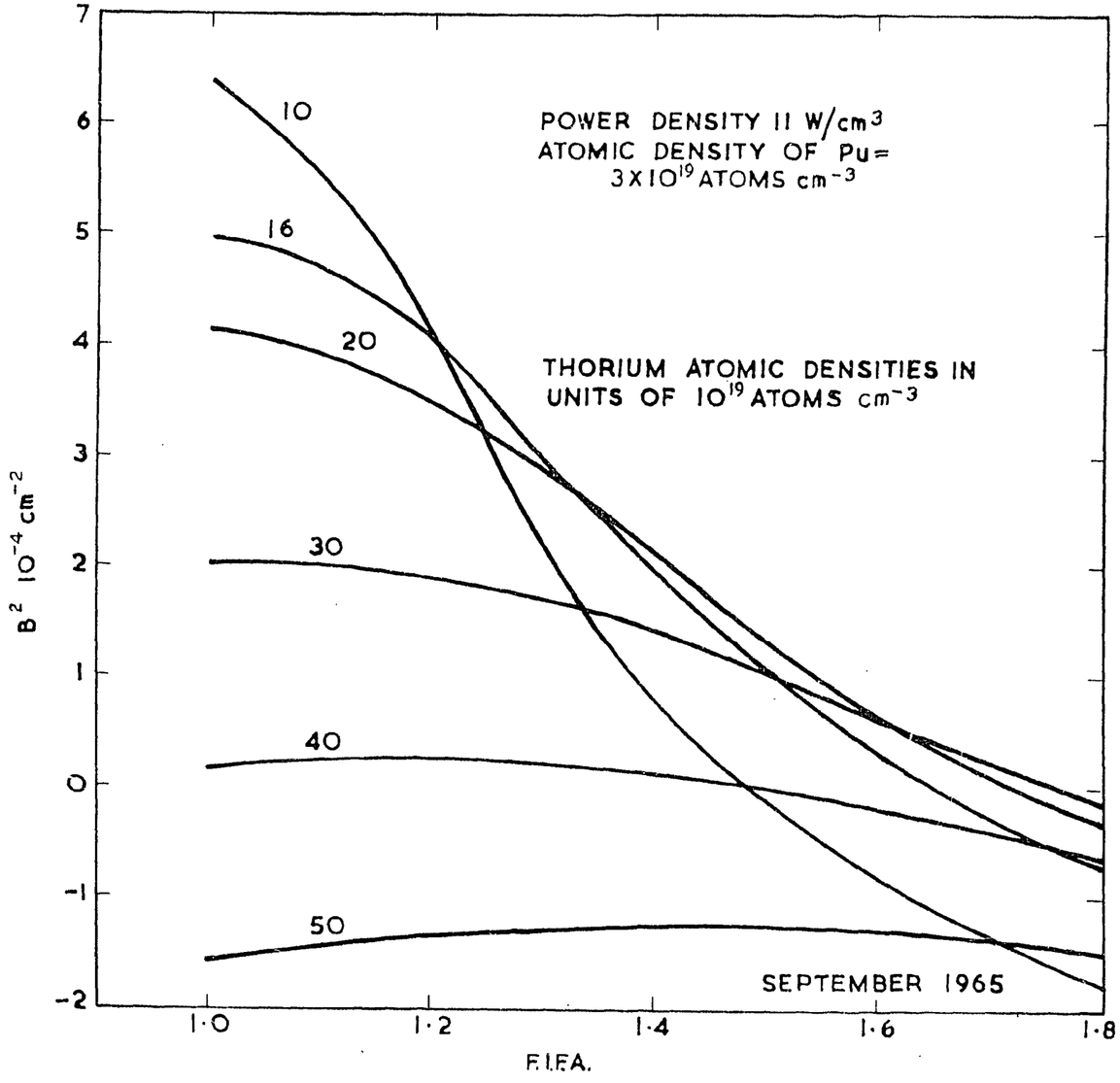

FIGURE 2. AVERAGE MATERIAL BUCKLING v. F.I.F.A.

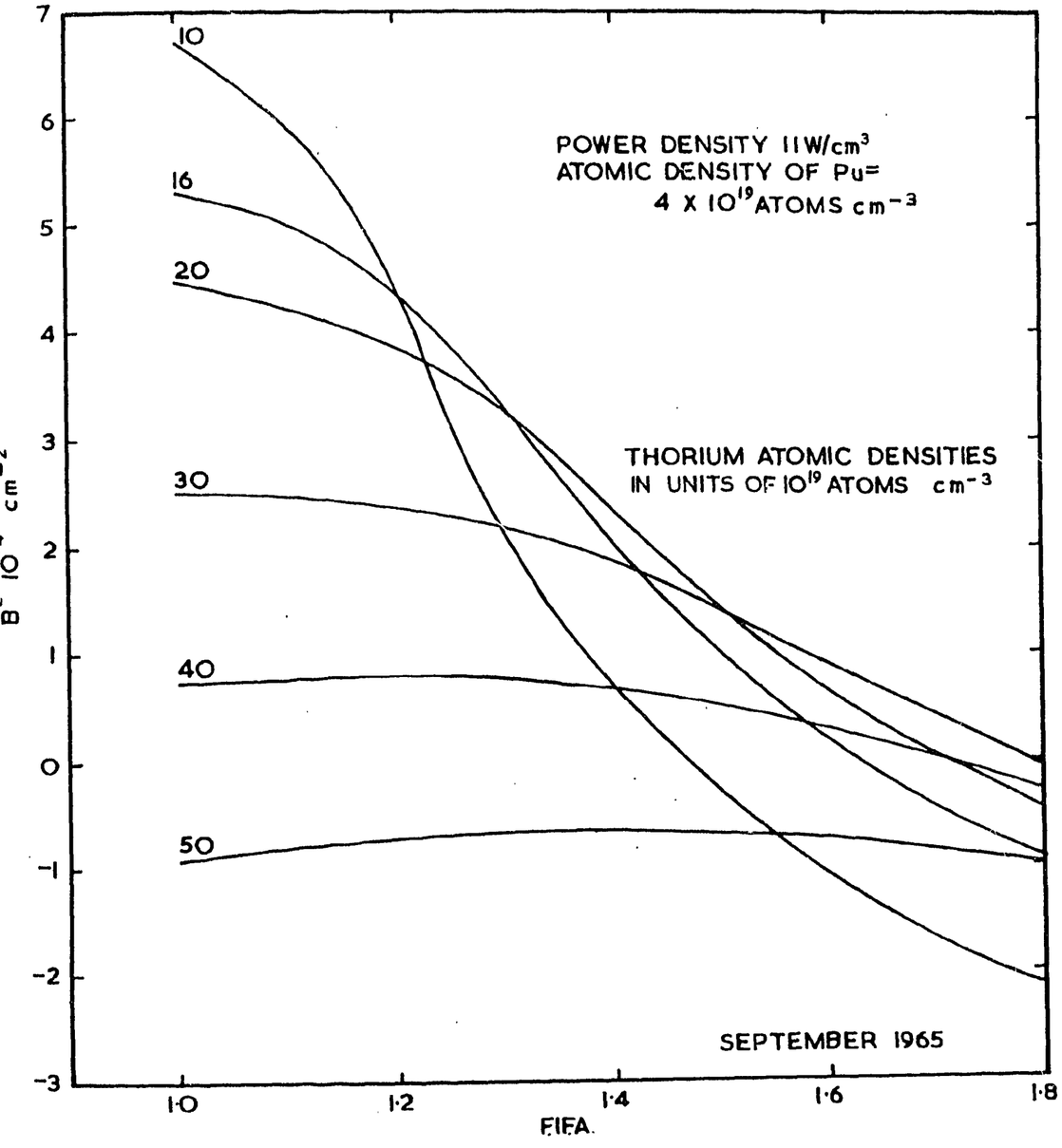

FIGURE 3. AVERAGE MATERIAL BUCKLING v. F.I.F.A.

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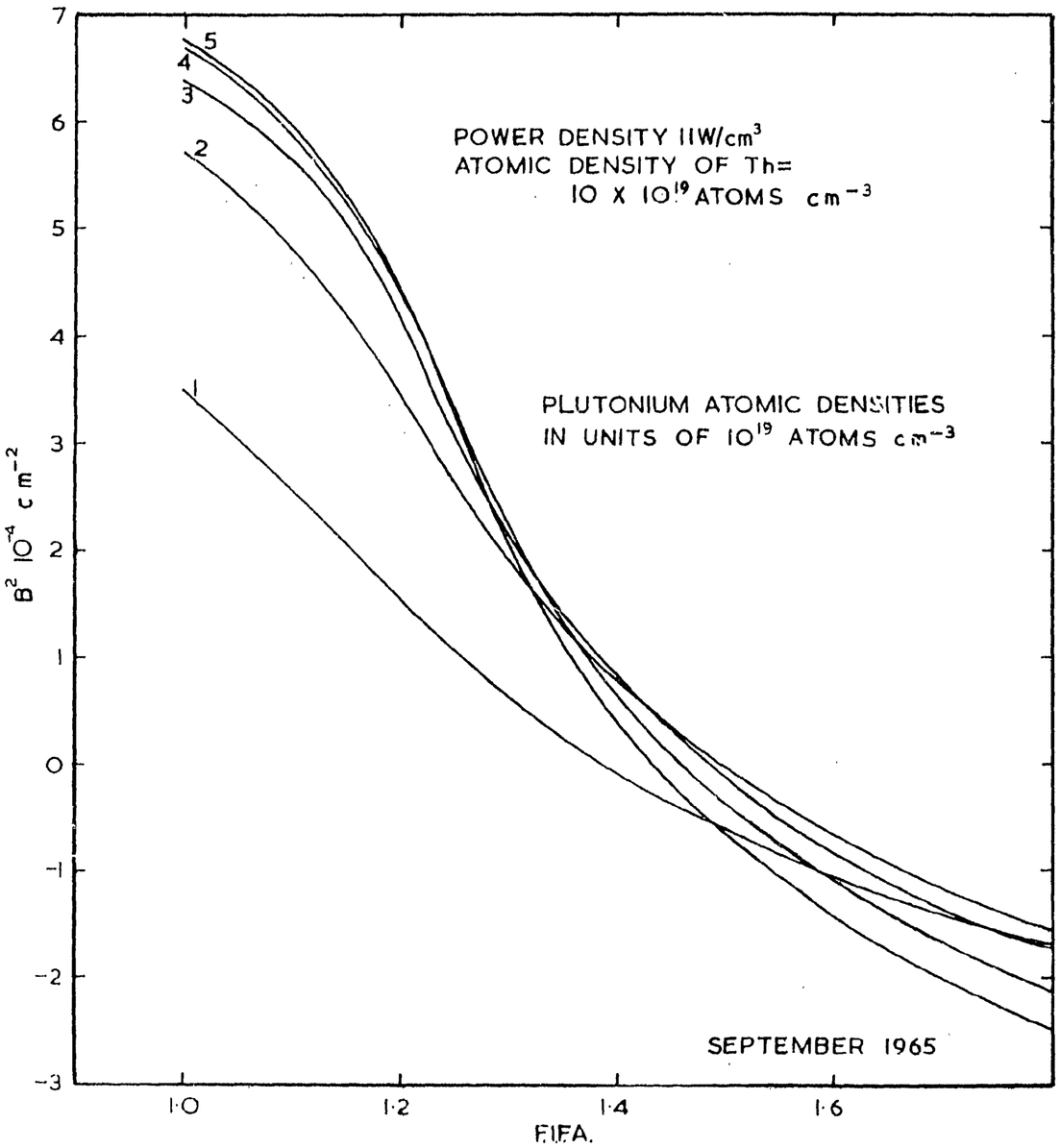

FIGURE 4. AVERAGE MATERIAL BUCKLING v. F.I.F.A.

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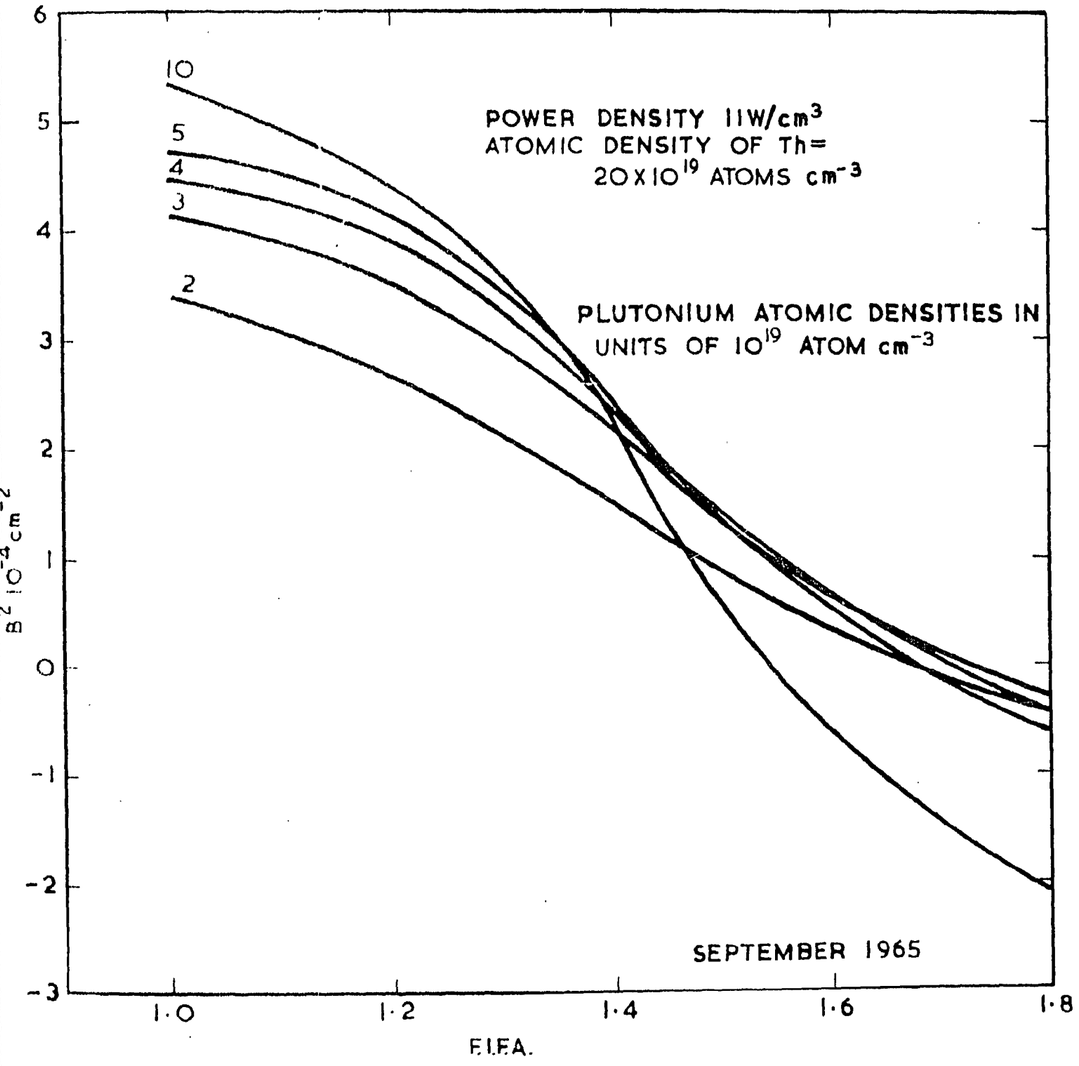

FIGURE 5. AVERAGE MATERIAL BUCKLING v. F.I.F.A.

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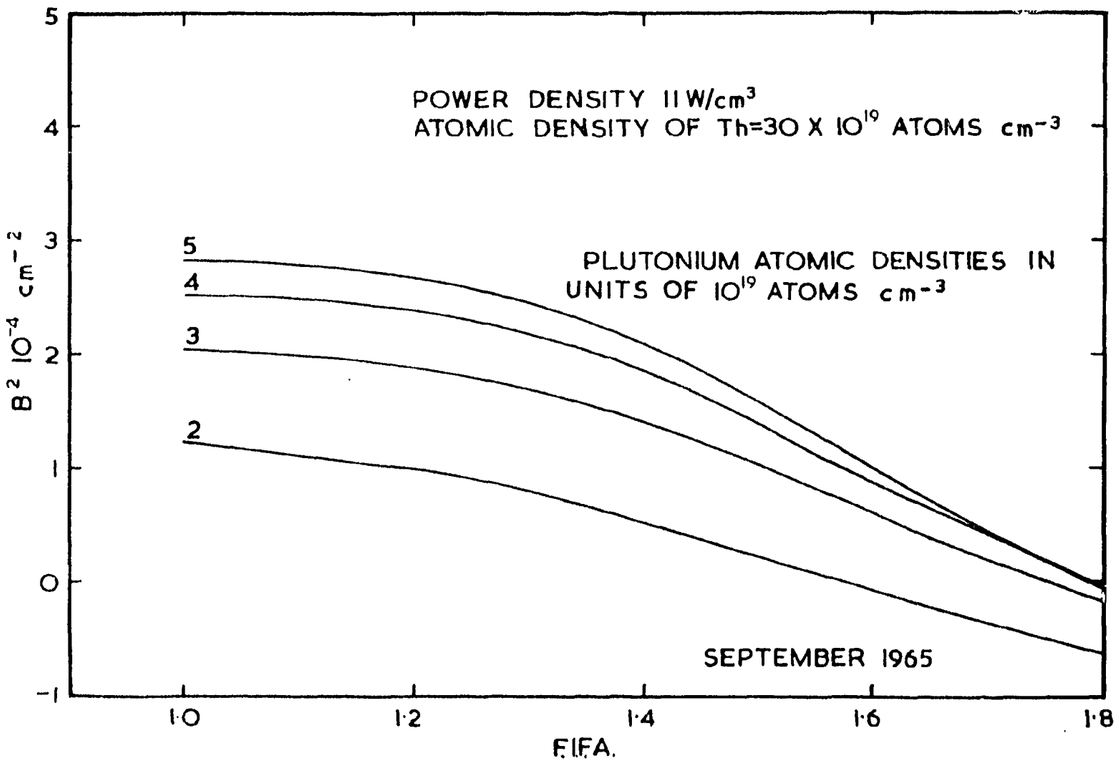

FIGURE 6. AVERAGE MATERIAL BUCKLING v. F.I.F.A.

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