Technical Report No. 32-198

A Parametric Survey of Criticality-Limited Fast Reactors Employing Uranium Fluoride Fuels

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ABSTRACT

Multigroup diffusion theory calculations are employed to estimate the size and weight of a uranium fluoride–fueled reactor. Beryllium, beryllium oxide, and graphite are investigated as possible reflector materials; diluents considered for the 93.5%-enriched fuel are the fluorides of sodium, lithium, beryllium, and zirconium. All survey calculations utilize the one-dimensional AIM-5 diffusion theory code (Ref. 1) and employ twelve energy groups.

It is estimated that for spacecraft reactors in the 10-Mw(th) class, the low fuel density of uranium fluoride results in reactors which are somewhat larger than comparable uranium carbide–fueled reactors. This assumes, however, that the solid fuel can achieve an average fuel burn-up of 30,000 Mwd/t, a performance which remains to be demonstrated for the 2000°F+ fuel-element surface temperatures that should characterize such reactors.

I. INTRODUCTION

Recent mission studies (Ref. 2, 3, 4) have shown that nuclear-electric spacecraft will be required to explore much of our planetary system. Since these spacecraft will be utilized to obtain information from distant regions of our solar system for extended periods of time, relatively long-lived nuclear reactors will be required as their energy source. A demand for reactors capable of supplying 10 Mw(th) power, or more, for time periods on the order of two to three years must be anticipated. Clearly, when 8,000-Mwd energy expenditures are cited for necessarily lightweight, compact reactors, fuel burn-up becomes a critical design consideration. The burn-up problem is aggravated further in this case because fuel temperatures will be high, but possibly not high enough for complete fission gas release. (Fuel surface temperatures which approach or exceed the 1100°C melting temperature of uranium metal can be predicted since heat-rejection temperatures in the order of 700 to 800°C will be required to obtain systems of reasonable size and weight). It is possible that the primary burn-up limitations of solid fuels (Ref. 5) can be circumvented by the use of liquid fuels if provisions are made for the removal of undissolved gaseous fission products. For this reason, it is instructive to compare the weight of a fluid-fueled reactor with that of a reactor which employs conventional solid-fuel elements, assuming that a total energy output of, say, 8,000 Mwd is required of each at 10 Mw (th) power.

As a liquid fuel, molten uranium fluoride is worthy of study: the physical and chemical properties of uranium fluoride are well reported (Ref. 6, 7) and its vapor pressure, unlike that of uranium chloride, is reasonable at the anticipated fuel temperatures. A substantial prompt negative temperature coefficient also can be anticipated for molten uranium fluoride–fueled reactors, since fuel expansion can be made essentially unrestricted. Although the low fuel density of uranium fluoride should result in somewhat larger criticality-limited reactors than many of the "conventional" solid fuels (uranium metal, uranium carbide, uranium dioxide, etc.), the magnitude of this differential may be reduced significantly if solid fuels cannot meet the burn-up requirements demanded by long-lived spacecraft power reactors.

Of the solid fuels which are capable of withstanding high temperatures, uranium carbide appears promising. Fuel burn-up in excess of 14,000 Mwd/t has been established for uranium carbide, although values as high as 20,000 Mwd/t remain to be demonstrated (Ref. 8). Uranium carbide is substantially superior to uranium dioxide as far as thermal conductivity is concerned (Ref. 8), and it is not clear that higher burn-up can be expected of the latter; thus, for the subsequent comparison with uranium fluoride, uranium carbide appears to be a reasonable selection.

II. ACCURACY OF THE CALCULATIONS

Twelve group cross sections were used to obtain the numerical results which appear in this Report. These cross sections were obtained or developed from data presented in Ref. 9 and 10. The utility of the uranium cross sections for predicting the critical mass of small reactors employing highly enriched fuel can be estimated from Table 1, in which the critical mass of a bare, spherical core of 93.5%-enriched uranium is shown to be 42.2 kg of uranium-235. This composition and configuration closely approximates that of the Godiva critical assembly (Ref. 9, 10), an assembly which is known to have a critical mass of 48.7 kg of uranium-235. Thus, the calculated critical mass of the bare uranium core shown in Table 1 is 12.7% too low. The corresponding error in critical radius is about 3.5%. No attempt was made to adjust the uranium cross sections or the number of neutrons produced per fission in order to obtain better agreement for this case.

The AIM-5 multigroup diffusion theory code (Ref. 1) was employed to obtain the results which follow in Section III; a very brief description of the equations solved by this code is given in the Appendix.

III. DISCUSSION OF RESULTS

Table 1 compares the steady-state properties of 93.5%-enriched uranium, uranium carbide, and uranium fluoride fuels, both bare and reflected with 3.0 in. (7.6 cm) of beryllium. As expected, the uranium carbide core is significantly smaller, yet heavier, than the uranium fluoride core for the bare case. The effect of fuel dilution becomes apparent, however, when the two fuels are reflected with 3.0 in. of beryllium, since the light reflector permits a substantial relative weight saving for the uranium carbide–fueled system. It should be pointed out that the uranium fluoride density shown in Table 1 is conservative: Oak Ridge National Laboratory has found the density of uranium fluoride to be 6.9 g/cm³ at 1100°C.[1]

Figures 1 through 6 and Table 2 present the results of a reflector survey for a uranium fluoride–fueled core composed of 60% fuel, 30% coolant and 10% structure by volume. Structural and heat-transfer analyses indicate that these percentages are reasonable for a high-power-density spacecraft reactor. Lithium was selected as the reactor coolant on the basis of its desirable heat-transfer characteristics, and zirconium as the reflector material because of its similarity to niobium (columbium), as far as nuclear properties are concerned.

A significant reactor weight saving can be realized by reflecting the 60% uranium fluoride, 30% lithium, 10% zirconium core with about 3 in. of beryllium, as shown in Fig. 1. Figure 2 shows, however, that reactor size increases monotonically with increasing reflector thickness for all three reflector materials. It is therefore clearly possible (if not likely) that the weight saving shown in Fig. 1 for a 3-in. beryllium reflector might be offset by weight change associated with a 3-in. change in the diameter of the payload shield. The usual critical mass decrease with increasing reflector thickness is shown in Fig. 3.

Spatial uniformity of the rate at which heat is generated in the core of a power reactor is highly desirable. This is particularly true for high-power-density reactors which utilize very high coolant temperatures. An indication of the power-distribution flattening that can be accomplished for a reactor core consisting of 60% uranium fluoride, 30% lithium, and 10% zirconium simply by reflecting it with beryllium, beryllium oxide, and graphite is provided by Fig. 4. This Figure presents two power-generation ratios, core-center-to-average and core-surface-to-average, as a function of reflector thickness. The two curves for each reflector material obviously intersect at the reflector thickness for which the maximum-to-average power-generation ratio is a minimum. Since a circulating fuel reactor cannot utilize a variable fuel loading for power-distribution flattening without considerable difficulty, the spherical-geometry calculations indicate that a maximum-to-average power-generation ratio of about 1.3 must be tolerated in such reactors if internal reflectors or islands are to be avoided. As seen from Fig. 4, a 6-in. graphite reflector is required to yield the same maximum-to-average power-generation ratio that can be obtained with 3 in. of beryllium or beryllium oxide. Plainly, a graphite-reflected reactor is too large (Fig. 2) and too heavy (Fig. 1) when compared to beryllium- or beryllium oxide–reflected cores of similar core spectra. The detailed spatial variation of the power generated per unit of core volume is shown in Fig. 5 for the bare core and two beryllium reflector thicknesses (3 and 6 in.). An estimation of the reactor thermalization with increasing reflector thickness is provided by Fig. 6.

It is interesting to note that the core consisting of 60% uranium fluoride, 30% lithium, and 10% zirconium derives a substantial reactor weight saving from a 3-in. beryllium reflector, whereas this saving is practically insignificant for pure uranium fluoride, as indicated in Table 1. This behavior can be attributed primarily to the relatively large transport mean free path of the lithium coolant.

Although beryllium oxide and graphite should exhibit satisfactory dimensional stability at high temperature, neutron-capture helium production may cause excessive swelling in pure beryllium if reflector temperatures exceed 700°C (Ref. 11).

Since the melting temperature of uranium fluoride is approximately 1050°C, some fuel dilution will be required to depress its melting temperature. Several diluents for uranium fluoride have been proposed and studied (Ref. 6). When examination is made of fuel density, melting temperature, vapor pressure, thermal conductivity, etc., it appears that a fuel consisting of 70% uranium fluoride and 30% sodium fluoride is about the best choice of the reported systems. A spherical reactor employing

[1] Private communication.

Table 1. Steady-state properties of enriched U, UC, and UF_4 fuels — spherical geometry

Item		Bare U core	Bare UC core	Bare UF_4 core	UC core reflected with 7.6 cm (3.0 in.) of Be		UF_4 core reflected with 7.6 cm (3.0 in.) of Be	
Core radius, cm		8.32	12.0	14.5	8.42		11.8	
Core volume, l		2.42	7.34	12.8	2.50		6.82	
Core density, g/cm^3		18.7	11.7	6.05	11.7		6.05	
Core weight, kg		45.2	85.9	77.4	29.2		41.3	
Critical mass, kg of U^{235}		42.2	76.7	54.7	26.0		29.2	
Reflector volume, l		—	—	—	14.8		23.7	
Reflector density, g/cm^3		—	—	—	1.85		1.85	
Reflector weight, kg		—	—	—	27.4		43.8	
Weight of core and reflector, kg		45.2	85.9	77.4	56.6		85.1	
Integrated fluxes								
Group	Energy interval				Core	Reflector	Core	Reflector
1	6.1 — ∞ Mev	0.0814	0.242	0.117	0.0835	0.114	0.182	0.133
2	3.7 — 6.1 Mev	0.396	1.07	0.591	0.423	0.597	0.831	0.646
3	1.4 — 3.7 Mev	1.74	5.48	2.74	2.09	3.29	4.42	3.47
4	0.50 — 1.4 Mev	1.89	6.21	2.98	2.40	3.18	5.11	3.11
5	0.18 — 0.50 Mev	0.994	3.74	1.70	1.58	2.52	3.26	2.33
6	3.4 — 180 kev	0.572	3.93	1.10	1.40	4.16	4.01	4.72
7	0.17 — 3.4 kev	0.001	0.132	0.142	0.0854	1.49	0.316	1.81
8	32 — 170 ev	—	—	—	0.0085	0.483	0.028	0.599
9	3.1 — 32 ev	—	—	—	0.0040	0.432	0.013	0.543
10	1.1 — 3.1 ev	—	—	—	0.0025	0.130	0.008	0.165
11	0.41 — 1.1 ev	—	—	—	0.0007	0.0958	0.003	0.123
12	0 — 0.41 ev	—	—	—	0.0009	0.266	0.003	0.350
Power-generation ratio, core center/average		1.89	2.06	2.57	1.22		1.56	
Power-generation ratio, core surface/average		0.517	0.444	0.238	1.38		1.14	
Neutrons produced by fission in GP 12, %		~0	~0	~0	0.905		1.33	
Neutrons escaping core/neutron produced		0.588	0.582	0.567	0.557		0.543	

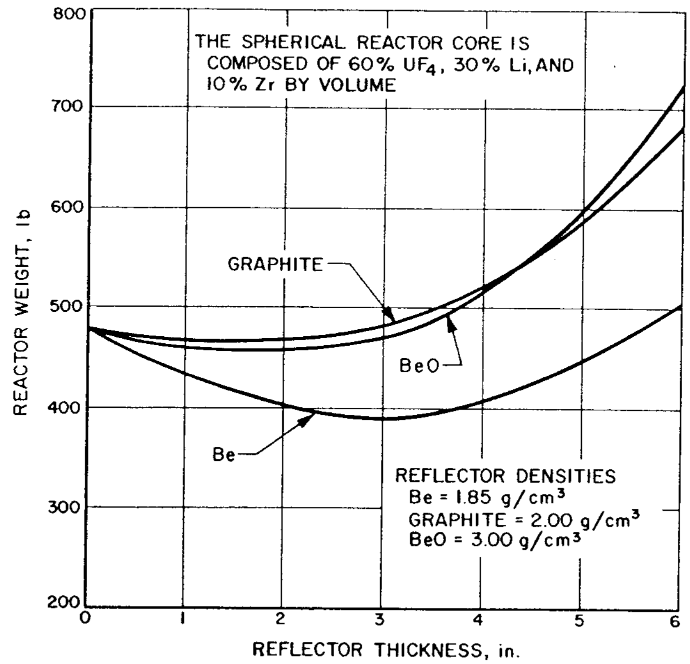

Fig. 1. Reactor weight vs reflector thickness for Be, BeO, and graphite reflectors

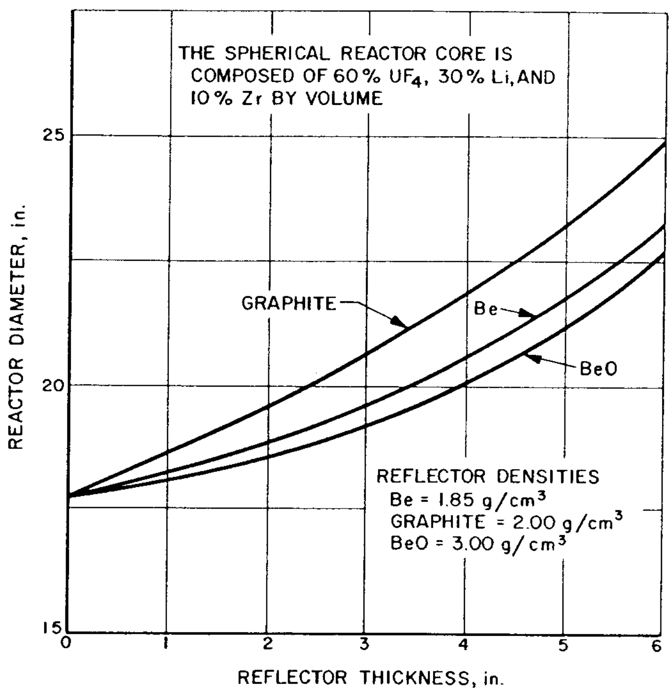

Fig. 2. Reactor diameter vs reflector thickness for Be, BeO, and graphite reflectors

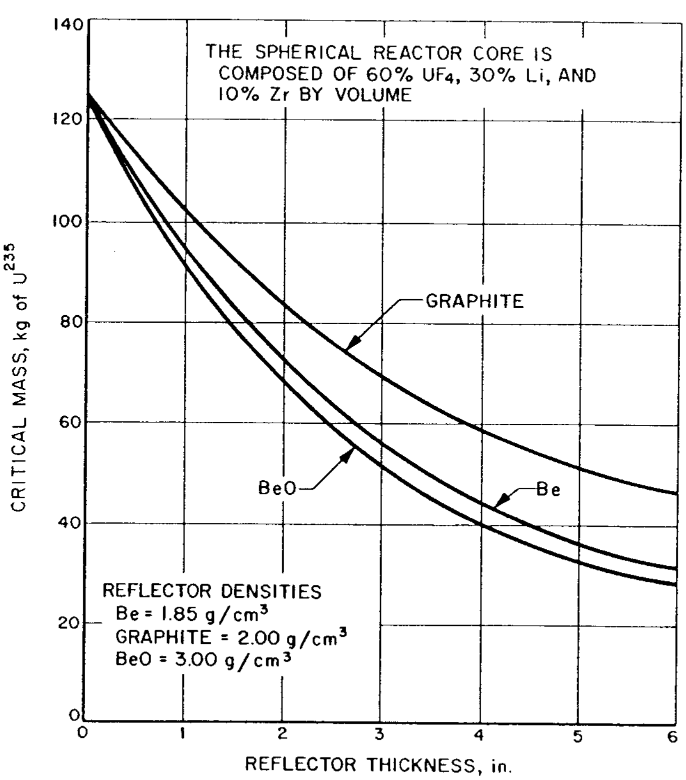

Fig. 3. Critical mass (U^{235}) vs reflector thickness for Be, BeO, and graphite reflectors

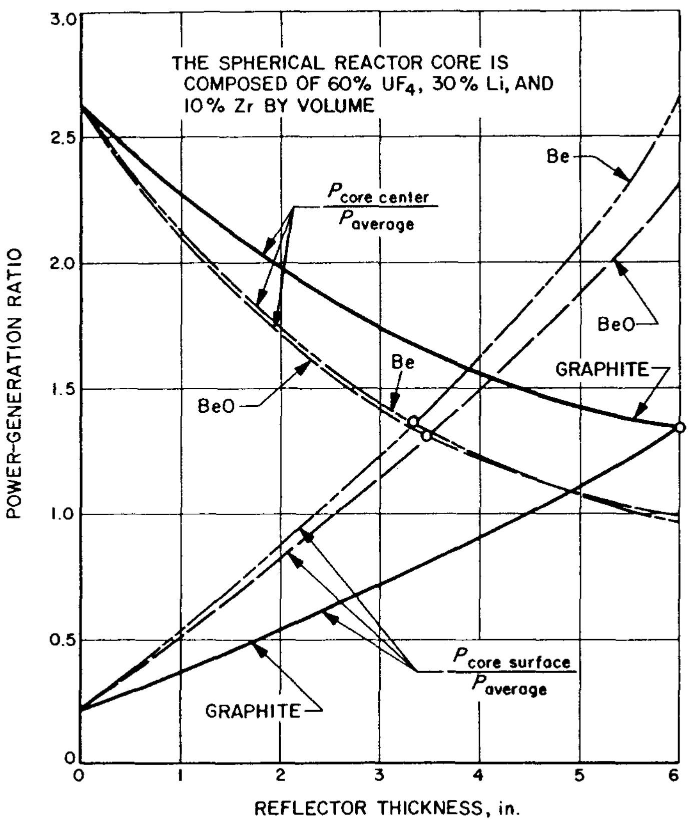

Fig. 4. Power-generation ratio vs reflector thickness for Be, BeO, and graphite reflectors

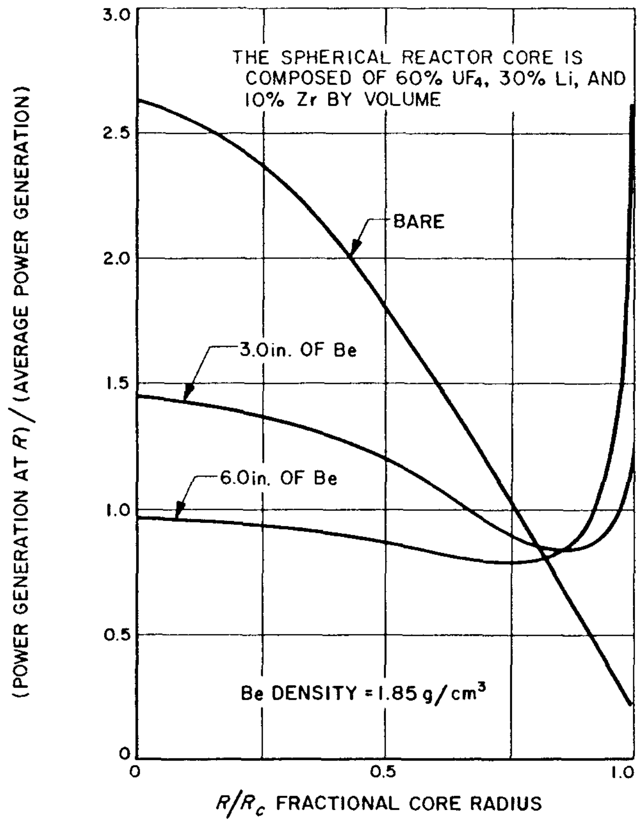

Fig. 5. Power-generation ratio vs core radius and reflector thickness

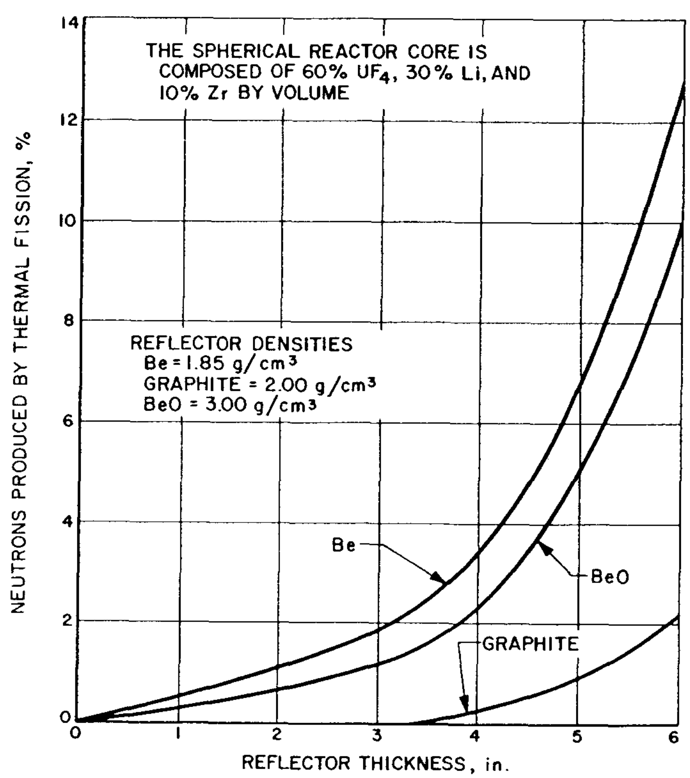

Fig. 6. Reactor thermalization vs reflector thickness for Be, BeO, and graphite reflectors

Table 2. Reflector comparison for a homogeneous, spherical core consisting of 60% UF_4, 30% Li, 10% Zr by volume

Item		Reflector thickness and material																
		Bare	7.6 cm of Be		7.6 cm of BeO		7.6 cm of C		7.6 cm of Ni		15.2 cm of Be		15.2 cm of BeO		15.2 cm of C		15.2 cm of Ni	
Core radius, cm		22.6	17.4		16.9		18.7		17.4		14.2		13.8		16.3		16.1	
Core volume, l		48.7	21.9		20.1		27.2		22.0		12.0		11.1		18.1		17.6	
Core density, g/cm^3		4.45	4.45		4.45		4.45		4.45		4.45		4.45		4.45		4.45	
Core weight, kg		217.	97.5		89.4		121.		97.9		53.4		49.4		80.5		78.3	
Critical mass, kg		125.	56.2		51.6		69.8		56.4		30.8		28.5		46.4		45.1	
Reflector volume, l		—	43.3		41.4		48.8		43.5		95.0		92.2		114.		112.	
Reflector density, g/cm^3		—	1.85		3.0		2.0		8.9		1.85		3.0		2.0		8.9	
Reflector weight, kg		—	80.1		124.0		97.6		387.		176.		277.		228.		997.	
Weight of core and reflector, kg		217.	177.6		213.		219.		485.		229.		326.		308.		1075.	
Integrated fluxes																		
Group	Energy interval		Core	Reflector	Core	Reflector	Core	Reflector	Core	Reflector	Core	Reflector	Core	Reflector	Core	Reflector	Core	Reflector
1	6.1 — ∞ Mev	0.351	0.262	0.124	0.245	0.107	0.281	0.115	0.274	0.088	0.203	0.202	0.188	0.162	0.236	0.192	0.254	0.107
2	3.7 — 6.1 Mev	1.61	1.24	0.619	1.21	0.601	1.37	0.636	1.25	0.378	0.966	1.05	0.950	0.973	1.18	1.15	1.16	0.452
3	1.4 — 3.7 Mev	8.63	6.82	3.53	6.71	3.62	7.71	3.58	7.31	3.39	5.35	6.70	5.33	6.72	6.74	7.07	6.85	5.29
4	0.50 — 1.4 Mev	10.0	7.95	3.24	7.97	3.27	9.02	3.72	9.28	5.55	6.11	5.80	6.24	5.58	7.80	7.59	9.05	12.1
5	0.18 — 0.50 Mev	6.03	5.07	2.46	5.11	2.58	5.62	2.45	5.73	3.41	3.94	4.76	4.05	4.70	4.92	5.67	5.70	8.67
6	3.4 — 180 kev	6.94	6.82	5.18	7.03	5.93	7.16	4.36	7.41	4.08	5.57	12.6	5.91	14.3	6.71	13.7	7.40	9.78
7	0.17 — 3.4 kev	0.286	0.630	2.02	0.649	2.14	0.486	1.09	0.473	1.22	0.701	6.97	0.744	7.74	0.655	5.84	0.539	4.42
8	32 — 170 ev	0.005	0.058	0.669	0.058	0.664	0.028	0.243	0.022	0.287	0.089	2.97	0.092	3.23	0.065	2.06	0.033	1.46
9	3.1 — 32 ev	0	0.028	0.609	0.025	0.545	0.007	0.128	0.004	0.143	0.056	3.95	0.056	4.11	0.032	2.00	0.009	1.05
10	1.1 — 3.1 ev	0	0.016	0.186	0.014	0.157	0.003	0.028	0.001	0.024	0.037	1.45	0.037	1.48	0.019	0.626	0.003	0.211
11	0.41 — 1.1 ev	0	0.006	0.139	0.005	0.110	0.001	0.015	0	0.009	0.015	1.31	0.015	1.30	0.007	0.485	0.001	0.087
12	0 — 0.41 ev	0	0.007	0.396	0.005	0.239	0.000	0.016	0	0.002	0.054	12.1	0.042	9.07	0.010	1.55	0	0.026
Power-generation ratio, core center/average		2.63	1.45		1.41		1.74		1.59		0.968		0.978		1.33		1.44	
Power-generation ratio, core surface/average		0.216	1.24		1.15		0.725		0.743		2.63		2.31		1.35		0.857	
Neutrons produced by fission in GP 12, %		~0	1.67		1.05		~0		~0		12.6		9.80		2.20		~0	
Neutrons escaping core/neutron produced		0.551	0.524		0.524		0.538		0.538		0.504		0.504		0.521		0.534	

this fuel is shown in Table 3. High-temperature corrosion and mass-transfer considerations will unquestionably be a large factor in the diluent selection; therefore, the fluorides of beryllium, lithium, and zirconium cannot be dismissed at this time.

It should be mentioned that fuel burn-up experiments conducted at Oak Ridge National Laboratory have not detected net cation or anion production as the result of uranium fluoride fission,[2] although previous theoretical work by ORNL indicated a possible excess of free fluorine. It is further anticipated that very high burn-up can be accomplished without gross fission product precipitation.

[2] Private communication.

IV. CONCLUSIONS

Assuming that an average burn-up of 30,000 Mwd/t can be achieved with uranium carbide fuel elements, a cylindrical core composed of 60% uranium carbide, 30% lithium, and 10% zirconium will weigh approximately 650 lb and occupy a volume of 34 l if the core is required to furnish an energy output of 8,000 Mwd. A beryllium reflector of 1.5-in. thickness (for reactor control) will add an additional 100 lb, resulting in a reactor weight of approximately 750 lb. Since reactor criticality demands a fuel enrichment of only 50 to 60%, and power densities of 500 kw/l appear reasonable for lithium-cooled reactors, an enriched uranium carbide–fueled reactor which weighs 750 lb and produces only 10-Mw heat is obviously burn-up limited.

If the reactor described in Table 3 is to be compared on a weight basis with the uranium carbide–fueled reactor described above, the critical mass shown in the Table must be adjusted to account for errors in the uranium cross sections and the shortcomings of diffusion theory. An increase of 13% will be assumed, although it is probably somewhat of an overestimate (Ref. 9). The transition from spherical to cylindrical geometry will require an additional critical mass increase of about 5% (Ref. 9), resulting in a reactor weight of about 685 lb for the cylindrical geometry. A heat-transfer analysis which was conducted concurrently with this criticality survey revealed that the low thermal conductivity of the fluoride fuels demands a circulating-fuel reactor design. This situation could not be rectified satisfactorily by extending the external surface of the coolant tubes; therefore, an additional weight increase of approximately 75 lb must be made to account for the fuel, fuel pump, and plumbing which are external to the core of a circulating fuel reactor and peculiar to that system. Fuel burn-up considerations demand an additional reactor weight increase on the order of 65 lb. Therefore, a cylindrical, circulating-fuel reactor, having a length-to-diameter ratio of unity and the core composition shown in Table 3, will weigh approximately 825 lb. At least 10 Mw(th) can be removed from the core of such a reactor if the lithium is pumped through tubes having an inside diameter of 3/8 in. at a heat flux of 750,000 Btu/ft^2-hr. Furthermore, an even greater power output potential exists since 1/4-in. tubes are not prohibitively small.

The preceding discussion indicates that the weight of a 10-Mw(th) spacecraft power plant will be increased very little if a criticality-limited, uranium fluoride–fueled reactor is substituted for a burn-up–limited, uranium carbide–fueled reactor. However, an additional weight penalty might arise from payload shielding considerations, since the diameter of the uranium fluoride–fueled reactor is about 25% larger. Even more significant is the fact that the reactor change does not result in a power plant that is definitely more reliable. The problems associated with high burn-up in solid fuels (fuel swelling, cracking, fuel-cladding separation, etc.) are replaced by the potential troubles of an additional liquid loop (pump cooling and lubrication, leaks, etc.). For these reasons it is felt that uranium fluoride does not achieve the full potential of liquid fuels. A static fuel which is effectively burn-up–

Table 3. Typical uranium fluoride–fueled fast reactor

(Spherical core composed of 42% UF_4, 18% NaF, 30% Li, and 10% Zr by volume, reflected with 3.0 in. of Be)

Item	Value
Core diameter, in.	21.0
Core volume, l	40.0
Core density, g/cm^3	3.6
Core weight, lb	312
Critical mass, kg of U^{235}	73
Reflector volume, l	61
Reflector density, g/cm^3	1.85
Reflector weight, lb	248
Reactor weight, lb	580
Fuel melting temperature, °F	~1250
Fuel vapor pressure at 2500°F, mm Hg	~100

Integrated fluxes

Group	Energy Interval	Core	Reflector
1	6.1 – ∞ Mev	0.323	0.117
2	3.7 – 6.1 Mev	1.52	0.588
3	1.4 – 3.7 Mev	8.55	3.42
4	0.5 – 1.4 Mev	10.3	3.20
5	0.18 – 0.5 Mev	6.75	2.47
6	3.4 – 180.0 kev	10.2	5.53
7	0.17 – 3.4 kev	1.09	2.22
8	32 – 170.0 ev	0.104	0.742
9	3.1 – 32.0 ev	0.047	0.680
10	1.1 – 3.1 ev	0.026	0.209
11	0.41 – 1.1 ev	0.010	0.157
12	0 – 0.41 ev	0.012	0.450

Power-generation ratio, core center/average	1.45
Power-generation ratio, core surface/average	1.25
Neutrons produced by fission in GP12, %	2.0
Median fission energy, kev	125

unlimited as far as radiation damage is concerned should be realizable in this fuel category.

If fuel-container corrosion rates at high temperatures prove acceptable, plutonium-based liquid fuels, such as the plutonium-iron eutectic under investigation at Los Alamos Scientific Laboratory, might provide a 10-Mw(th) space power reactor with its size determined solely by heat-transfer considerations. A "slurry" fuel consisting of uranium carbide pellets or granules and a liquid heat-transfer medium such as lithium also may achieve this objective. As conceived, the slurry would consist of 70 to 85% uranium carbide by volume, the remaining void being filled with the liquid heat-transfer medium.

The Jet Propulsion Laboratory is undertaking an experimental program to determine compatibility and fuel-container corrosion rates at temperatures of interest for some of the fluorides mentioned earlier, as well as the uranium carbide–lithium slurry. Several container materials, including columbium-, tantalum-, and tungsten-based alloys, will be investigated.

ACKNOWLEDGMENT

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APPENDIX

The AIM-5 Code

The AIM-5 code is a one-dimensional, multigroup diffusion theory code (Ref. 1) which solves the equations

$$D^i \nabla^2 \phi^i + \Sigma_T^i \phi^i = \chi^i S(r) + \sum_{j=q}^{i-1} \Sigma_{s,\, j \to i}\, \phi^j$$

where

D_i = diffusion coefficient for the ith group,

ϕ^i = neutron flux in the ith group,

Σ_T^i = total removal from the ith group,

$$\Sigma_T^i = \Sigma_a^i + \sum_{j=i+1}^{n} \Sigma_{s,\, i \to j} \,,$$

Σ_i^a = absorption cross section for the ith group,

$\Sigma_{s,\, i \to j}^i$ = scattering or transfer coefficient from group i to group j,

χ^i = the integral of the fission spectrum over the lethargy range represented by group i,

$$S(r) = \sum_i \left[\left(\nu^i \Sigma_f^i \right) \phi^i / \lambda \right],$$

r = distance measured from the origin,

Σ_f^i = fission cross section for the ith group,

ν^i = the average number of neutrons produced by a fission in the ith group, and

λ = the eigenvalue, which is related to the multiplication factor,

for as many as twelve energy groups. Neutron downscattering is permitted from any group to all lower groups.

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