RELATIVE NEUTRONIC PERFORMANCE OF PROPOSED HIGH-DENSITY DISPERSION FUELS IN WATER-MODERATED AND D₂O-REFLECTED RESEARCH REACTORS*

M. M. Bretscher, J. E. Matos, and J. L. Snelgrove

Argonne National Laboratory
Argonne, Illinois 60439-4841 USA

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Abstract

This paper provides an overview of the neutronic performance of an idealized research reactor using several high density LEU fuels that are being developed by the RERTR program. High-density LEU dispersion fuels are needed for new and existing high-performance research reactors and to extend the lifetime of fuel elements in other research reactors. This paper discusses the anticipated neutronic behavior of proposed advanced fuels containing dispersions of U$_3$Si$_2$, UN, U$_2$Mo and several uranium alloys with Mo, or Zr and Nb. These advanced fuels are ranked based on the results of equilibrium depletion calculations for a simplified reactor model having a small H$_2$O-cooled core and a D$_2$O reflector. Plans have been developed to fabricate and irradiate several uranium alloy dispersion fuels in order to test their stability and compatibility with the matrix material and to establish practical loading limits.

INTRODUCTION

Although many research reactors have been converted from HEU to LEU fuel since the beginning of the RERTR program,[1] conversion of high performance reactors awaits the development, testing, and licensing of high-density LEU fuels. At the present time the highest density dispersion fuel qualified for research reactor use is U$_3$Si$_2$-Al with a density of 4.8 gU/cm^3 in the fuel meat.[2] Recently Durand reported[3] that five U$_3$Si$_2$-Al MTR-type fuel plates each with a uranium loading of 6.0 g/cm^3 are being irradiated in the SILOE reactor and that this uranium density is a practical upper limit for U$_3$Si$_2$-Al fuel. CERCA has also fabricated[4] UN-Al MTR-type plates with a uranium density of 7.0 g/cm^3 and found that UN is compatible with the aluminum matrix at temperatures below 500°C. However, no irradiation tests of uranium mononitride dispersion fuel have been conducted.

The high-density LEU dispersion fuels proposed for development are based on alloys of uranium with elements known to stabilize the gamma phase. Irradiation tests to determine the combinations and the amounts of alloying additions needed are being prepared. This paper compares the performance of some of these high-density, uranium-alloy dispersion fuels with the performance of U$_3$Si$_2$-Al and UN-Al in an idealized MTR-type research reactor moderated with

light water and reflected with heavy water in order to identify those high-density fuels best suited for development from a neutronics point of view. Although this paper focuses on neutronic properties of these advanced high-density fuels, the primary requirement is to choose alloy compositions which are compatible with the matrix material, which stabilize the uranium by inhibiting the formation of the α-phase under intense irradiation conditions, and which satisfy requirements for licensing and final disposal.

PROPERTIES OF HIGH-DENSITY DISPERSION FUELS

The density of uranium in the fuel meat increases linearly with the volume fraction of the dispersant. Thus,

$$\rho_U = \rho^D \ W_U^D \ VF^D$$

where ρ_U is the uranium density in the fuel meat, ρ^D is the theoretical density (TD) of the dispersant, W_U^D is the weight fraction of uranium in the dispersant, and VF^D is the volume fraction of the dispersant in the fuel meat exclusive of dispersant internal voids. Table 1 gives these dispersant properties for the high-density fuels analyzed in this study. Note that for a given volume fraction ρ_U is largest for that dispersant for which the product $\rho^D W_U^D$ is a maximum.

The porosity (P) in the fuel meat depends on the volume fraction of the dispersant, other fuel properties such as the volume fraction of fine particles, and fabrication process variables.[5] For U_3Si_2 fuel fabricated at ANL the porosity is given by the relation[6]

$$P = 0.072 \ VF^D - 0.275(VF^D)^2 + 1.32(VF^D)^3.$$

This relation approximately accounts for the observed porosity of the UN-Al test plates reported in Ref. 4. The rapid increase in porosity at high volume fractions owes principally to the breakup of the brittle fuel particles. Since dispersants consisting of uranium alloys are less brittle than the U_3Si_2 and UN compounds, one expects the porosity to be smaller. Based on porosity measurements with uranium-tungsten alloys,[7] the porosity for the uranium alloys considered in this study was assumed to be one half of that predicted by the above equation.

The volume fraction of the aluminum matrix (VF^M) in the fuel meat is

$$VF^M = 1.0 - VF^D - P.$$

Table 2 summarizes the volume fractions and uranium densities for the high-density fuels analyzed in this study. From a practical point of view, it is unlikely that fuels can be fabricated with dispersant volume fractions above the 50-55% range. For the qualified U_3Si_2-Al fuel (4.8 gU/cm^3), $VF^D = 42.4\%$. The CERCA UN-Al (7.0 gU/cm^3) and U_3Si_2-Al (6.0 gU/cm^3) test plates have VF^D values of about 52% and 53%, respectively.

For all the cases considered in this study the LEU fuel had a ^{235}U enrichment of 19.89 w/o.

Table 1. Properties of Uranium Dispersants

Dispersant	ρ^D-g/cm^3	W_U^D	$\rho^D*W_U^D$
Compounds:			
U_3Si_2	12.2	0.927	11.3
UN	14.3	0.944	13.5
U_2Mo	16.6	0.832	13.8
U_2Ti	15.1	0.909	13.7
U_2Ru	16.9	0.825	13.9
Alloys:			
U-Mo(9 w/o)	17.0	0.910	15.5
U-Mo(5 w/o)	17.9	0.950	17.0
U-Zr(3 w/o)-Nb(9 w/o)	16.2	0.880	14.3
U-Zr(4 w/o)-Nb(2 w/o)	17.3	0.940	16.3

Table 2. Characteristics of the High-Density Uranium Dispersion Fuels

Dispersant	VF^D %	ρ_U g/cm^3	P %	VF^M %	M(^{235}U)/plate g
Compounds:					
U_3Si_2	42.4	4.80	8.2	49.4	17.65
U_3Si_2	50.0	5.66	13.2	36.8	20.79
UN	50.0	6.75	13.2	36.8	24.83
U_2Mo	50.0	6.90	13.2	36.8	25.39
Alloys:					
U-Mo(9 w/o)	42.4	6.57	4.1	53.5	24.15
U-Mo(9 w/o)	50.0	7.74	6.6	43.4	28.45
U-Mo(9 w/o)	57.5	8.90	8.0	34.5	32.72
U-Mo(5 w/o)	50.0	8.50	6.6	43.4	31.27
U-Zr(3 w/o)-Nb(9 w/o)	50.0	7.13	6.6	43.4	26.21
U-Zr(4 w/o)-Nb(2 w/o)	50.0	8.13	6.6	43.4	29.90

IDEALIZED REACTOR MODEL AND MULTIGROUP CROSS SECTIONS

A simplified reactor model was used for the purpose of this study. It consists of a 3x3 array of MTR-type fuel elements cooled with light water and reflected with heavy water. A beryllium block with a small water hole along its axis occupies the central location. Each of the 8 fuel elements contains 18 fuel plates with fuel meat and cladding thicknesses of 0.508 mm and 0.381 mm, respectively. The thickness of the coolant channel is 3.230 mm. Effective dimensions for the width and length of the fuel meat are 6.096 cm and 59.69 cm. Except for a thinner plate (1.27 mm rather than 1.52 mm), the geometry of the fuel element is identical to that used in the Ford Nuclear Reactor which is described in detail in Ref. 8. Table 2 shows the initial ^{235}U plate loading for each fuel composition. No control rods were modeled in this study.

The WIMS-D4M code[9] was used to generate 4-group, burnup-dependent, cross sections using an ENDF/B-V based 69-group library.[10] Multigroup cross sections were calculated for the active homogenized (fuel-clad-coolant) region, for the inactive side plate regions, and for the reflector regions. Cross sections were generated for each fuel type and for each uranium density. These cross sections were used with the REBUS code[11] to perform numerous equilibrium cycle depletion calculations. A fuel management scheme was adopted in which two fuel elements were added/discharged per cycle. For a reactor power of 10 MW, the cycle length was defined as the number of full power days (FPD's) required to give an eigenvalue at the end of the equilibrium cycle (EOEC) of 1.0150.

RESULTS

The results of the equilibrium fuel depletion calculations are summarized in Tables 3-5. Table 3 lists beginning of life (BOL), beginning of equilibrium cycle (BOEC), and end of equilibrium cycle (EOEC) eigenvalues for various fuel loadings and operating times. These data were used to determine the cycle length corresponding to k_{eff} (EOEC) = 1.0150 for each fuel type. This table includes results from calculations where the atom densities of the non-uranium part of the fuel dispersant were set equal to zero. These results allowed a determination of the cycle length loss resulting from neutron absorption in the diluent of the dispersant.

Table 4 shows the cycle lengths for each fuel loading calculated with (CL_p) and without (CL_o) parasitic absorption in Si, N, Zr, Nb, and Mo. At a dispersant volume fraction of 50%, the loss in cycle length from this parasitic absorption is 15.5% for UN, 10.2% for U-Mo (9%), 4.5% for U-Mo(5%), 3.6% for U-Zr(3%)-Nb(9%), 0.7% for U-Zr(4)-Nb(2%), and 0.2% for U_3Si_2. Whereas it is important to choose materials which minimize this parasitic absorption the most important single parameter influencing the cycle length is ρ_U, the uranium density in the fuel meat. Table 4 also includes the ^{235}U discharge burnups.

Figure 1 is a plot of the cycle length CL_o as a function of uranium density in the fuel meat. This graph shows that without neutron absorption in the fuel diluent, CL_o is a linear function of ρ_U. Cycle length decreases resulting from neutron absorption in the diluent are given in Table 4. These cycle length losses are plotted in Fig. 2 as a function of the diluent macroscopic absorption cross section, taken to be

Table 3. REBUS Equilibrium Cycle Eigenvalues

Fuel	VF^D %	ρ_U g/cm^3	FPD's	Eigenvalues		
				BOL	BOEC	EOEC
U$_3$Si$_2$	42.4	4.80	10	1.10969	1.04961	1.02069
U$_3$Si$_2$	42.4	4.80	12	1.10969	1.04496	1.01261
U*	42.4	4.80	12	1.10986	1.04512	1.01278
U$_3$Si$_2$	50.0	5.65	19	1.13553	1.05934	1.01921
U$_3$Si$_2$	50.0	5.65	20	1.13553	1.05724	1.01549
U*	50.0	5.65	20	1.13563	1.05735	1.01563
UN	50.0	6.75	25	1.14350	1.06289	1.01964
UN	50.0	6.75	26	1.14350	1.06109	1.01645
U*	50.0	6.75	26	1.15735	1.07439	1.02974
U$_2$Mo	50.0	6.90	25.0	1.13815	1.05904	1.01697
U$_2$Mo	50.0	6.90	25.6	1.13815	1.05801	1.01515
U*	50.0	6.90	25.6	1.15974	1.07836	1.03500
U-Zr(3%)-Nb(9%)	50.0	7.13	29	1.15979	1.07410	1.02744
U-Zr(3%)-Nb(9%)	50.0	7.13	33	1.15979	1.06702	1.01486
U*	50.0	7.13	33	1.16407	1.07106	1.01884
U-Zr(4%)-Nb(2%)	50.0	8.13	40	1.17681	1.07941	1.02424
U-Zr(4%)-Nb(2%)	50.0	8.13	43	1.17681	1.07447	1.01541
U*	50.0	8.13	43	1.17776	1.07536	1.01630
U-Mo(9%)	42.4	6.57	22	1.14337	1.06707	1.02719
U-Mo(9%)	42.4	6.57	26	1.14337	1.05980	1.01432
U*	42.4	6.57	26	1.15408	1.06989	1.02416
U-Mo(9%)	50.0	7.74	33	1.16057	1.07245	1.02408
U-Mo(9%)	50.0	7.74	36	1.16057	1.06754	1.01538
U*	50.0	7.74	36	1.17235	1.07860	1.02612
U-Mo(9%)	57.5	8.90	46	1.17264	1.07220	1.01482
U-Mo(9%)	57.5	8.90	45	1.17264	1.07369	1.01749
U*	57.5	8.90	45	1.18553	1.08576	1.02918
U-Mo(5%)	50.0	8.50	44	1.17509	1.07446	1.01684
U-Mo(5%)	50.0	8.50	45	1.17509	1.07287	1.01400
U*	50.0	8.50	45	1.18163	1.07899	1.01992

* The non-uranium component of the dispersant was omitted in these calculations
  in order to determine the effect of neutron absorption in the fuel diluent.

Table 4. Cycle Lengths and Discharge Burnups

Fuel	VF^D %	ρ_U g/cm^3	$-W_p^a$ %δk/k	Cycle Length,[b] FPD's			Discharge BU,%
				CL_p	CL_o	% Loss	
Compounds:							
U_3Si_2	42.4	4.80	0.017	11.41	11.45	0.35	23.7
U_3Si_2	50.0	5.65	0.014	20.13	20.17	0.20	32.3
UN	50.0	6.75	1.270	26.5	30.6	15.47	36.1
U_2Mo	50.0	6.90	1.889	25.6	32.1	25.39	34.8
Alloys:							
U-Zr(3%)-Nb(9%)	50.0	7.13	0.385	33.0	34.2	3.64	43.1
U-Zr(4%)-Nb(2%)	50.0	8.13	0.086	43.1	43.4	0.70	48.8
U-Mo(9%)	42.4	6.57	0.947	25.8	28.9	12.02	37.1
U-Mo(9%)	50.0	7.74	1.031	36.1	39.8	10.25	43.3
U-Mo(9%)	57.5	8.90	1.116	45.9	50.2	9.42	47.8
U-Mo(5%)	50.0	8.50	0.572	44.6	46.7	4.71	48.8

[a] W_p is the reactivity worth of the fuel diluent at EOEC.

[b] The cycle length (CL) is the number of 10 MW Days (FPD's) needed to produce an eigenvalue of 1.0150 at the end of the equilibrium cycle. CL_p and CL_o are cycle lengths with and without neutron absorption in the fuel diluent.
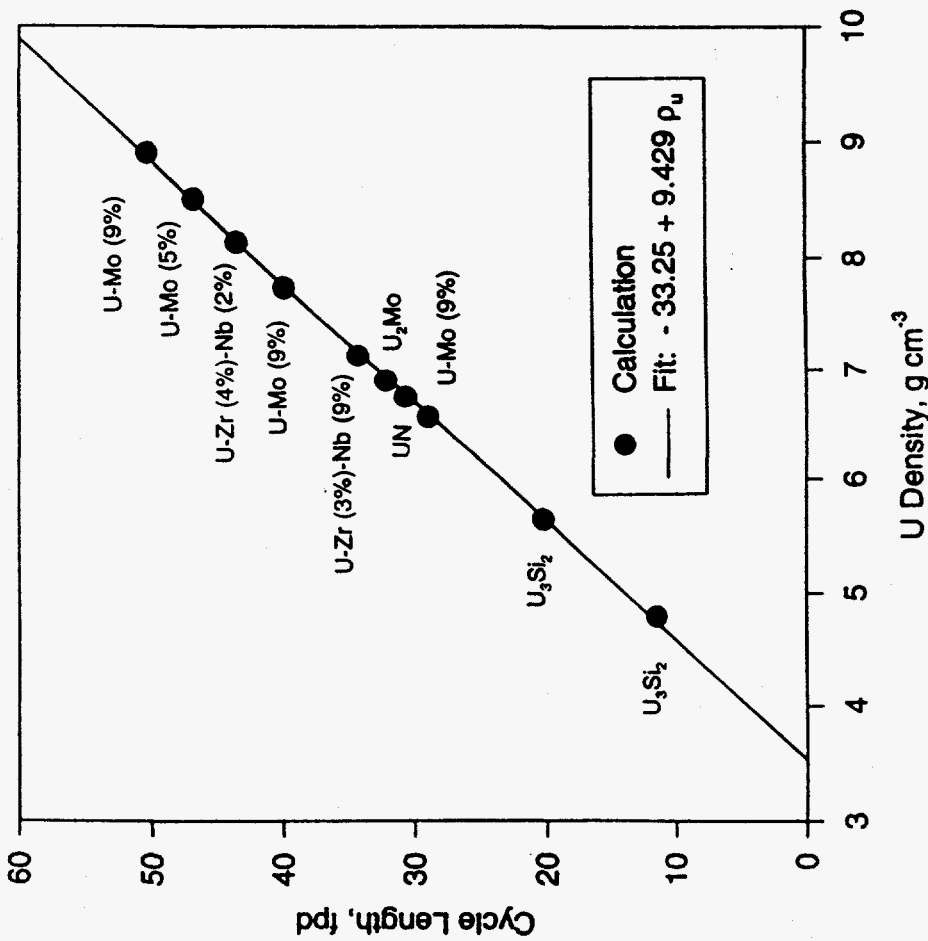
Figure 1. Cycle Length (CL_o) vs. U Density for 3 x 3 Core Without Fuel Diluent
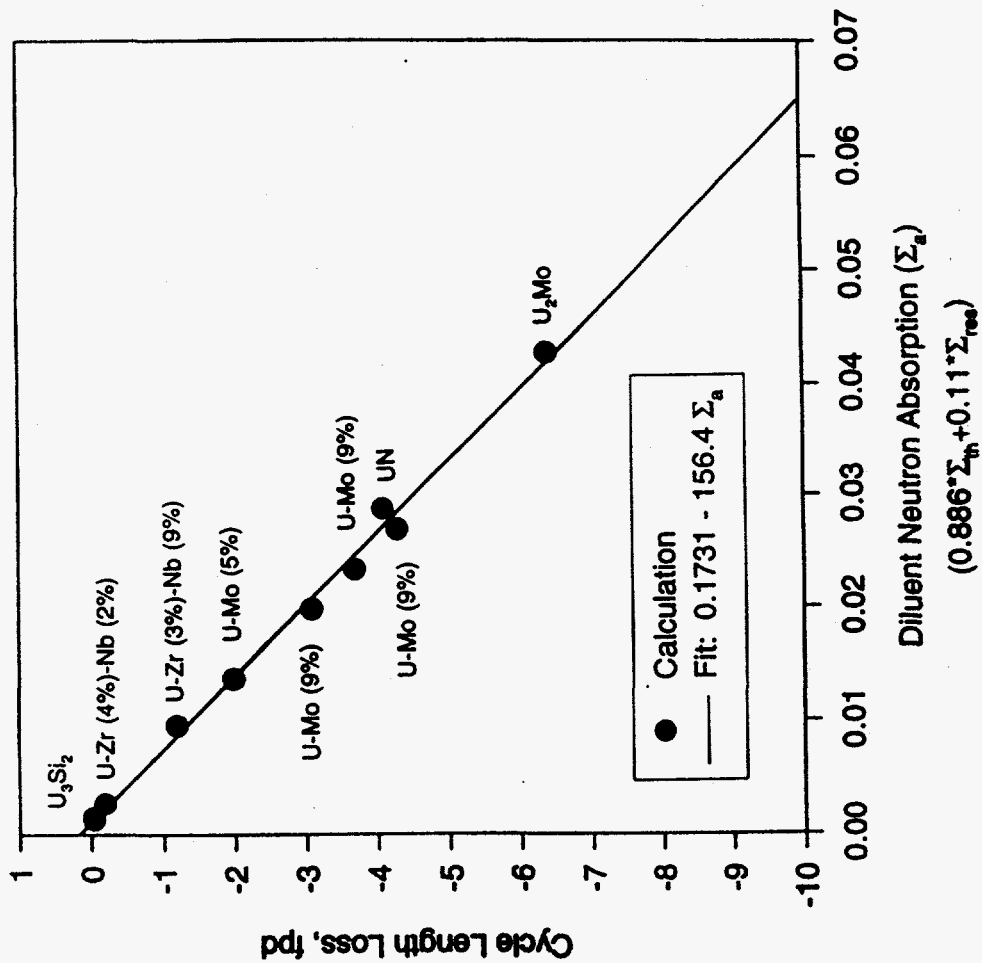
Figure 2. Cycle Length Loss (ΔCL) as a Function of Diluent Neutron Absorption

$$\Sigma_a = N_D\left(\frac{\sqrt{\pi}}{2}\sigma_{th} + \frac{RI}{\ln(5530/0.625)}\right)$$

where N_D is the diluent atom density in the fuel meat, σ_{th} is the thermal neutron absorption cross section of the diluent, and RI is its resonance integral. The argument of the logarithm is the ratio of the energy boundaries of the resonance group (Gp 3). If the thermal and resonance group fluxes in the core are nearly equal, Σ_a is proportional to the diluent absorption rate. Figure 2 shows that the cycle length loss, ΔCL, is well fit by a linear function of Σ_a. This correlation, together with Fig. 1, allows one to estimate the cycle length of any proposed advanced dispersion fuel in the 3x3 core without the need to perform detailed depletion calculations. For example, a dispersion of the compound U_2Ti at $VF^D = 50\%$ has a value of $\rho_U = 6.86$ g/cm^3 (see Table 1), $N_D(Ti) = 8.682E\text{-}3/b\text{-}cm$, and $\Sigma_a(Ti) = 0.050$ cm^{-1}. The correlations in Figs. 1 and 2 predict a cycle length of 23.8 FPD's. Similarly, the predicted cycle length for a dispersion of U_2Ru at $VF^D=50\%$ is 23.3 FPD's.

These correlations may be used to determine the cycle length (CL_p) for each dispersant as a function of ρ_U or VF^D. Since

$$CL_P = CL_o + \Delta CL = -33.08 + 9.429\,\rho_u - 156.4\,\Sigma_a$$

and since Σ_a is proportional to ρ_U, the cycle lengths are linear functions of ρ_U or VF^D. They are plotted in Figures 3 and 4. Figure 3 shows that for a fixed value of the uranium density the longest cycle length is obtained for the dispersant whose diluent is least absorbing, which is U_3Si_2. UN has a shorter cycle length because of parasitic loss from the $^{14}N(n,p)$ process. On the other hand, Fig. 4 shows that for a fixed value of the dispersant volume fraction UN has a longer cycle length than U_3Si_2. Thus, ranking of the advanced dispersion fuels by cycle length depends on whether the uranium density is held constant (Fig. 3) or whether the dispersant volume fraction is held constant (Fig. 4).

If there are internal voids within the dispersant, the dispersant density is $(1-P_i)\rho^D$ and the dispersant volume is $VF^D/(1-P_i)$, where P_i is the internal porosity. Except for UN, the uranium alloys and compounds considered in this study have essentially zero P_i values. Reference 4 reports that the uranium nitride powder used to fabricate UN-Al fuel plates had an internal porosity of about 10%. With this P_i value, the UN plot shown in Fig. 4 is displaced to the right and becomes nearly co-linear with the U_3Si_2 plot.

Midplane neutron fluxes at the beginning of the equilibrium cycle (BOEC) and at a power level of 10 MW are given in Table 5. They refer to a small H_2O hole at the center of the beryllium block and a D_2O region 5.0 cm thick surrounding the core. Effects of spectral hardening and flux reduction with increasing uranium density are evident. Figure 5 is a plot of the D_2O thermal neutron flux ($E_n<0.625$ eV) as a function of uranium density using data given in Table 5. The curve is drawn through the U_3Si_2 and U-Zr(4%)-Nb(2%) data points for which parasitic absorption in the fuel diluent is negligible. Points below this line show that neutron capture by the dispersant diluent reduces the D_2O thermal neutron flux. Since the product of k_{eff} and the thermal neutron flux is nearly independent of control rod elevation, this product is an effective indication of the actual flux at critical.
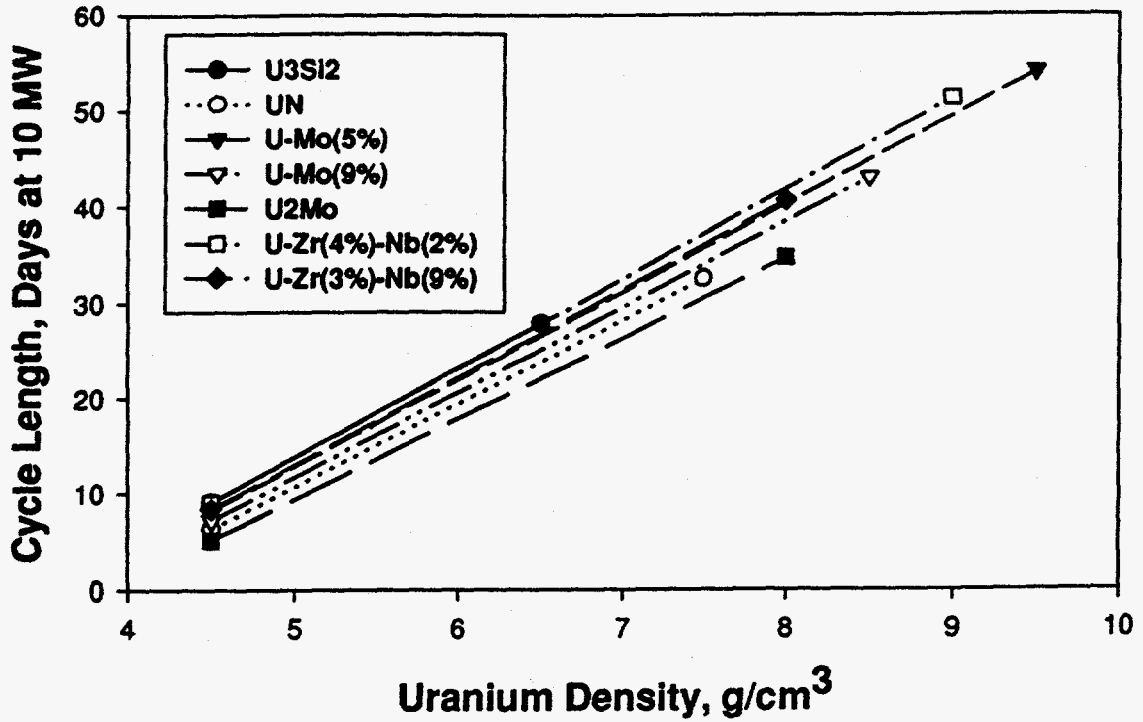
Figure 3. Cycle Length vs Uranium Density for Advanced Fuels in 3x3 Core
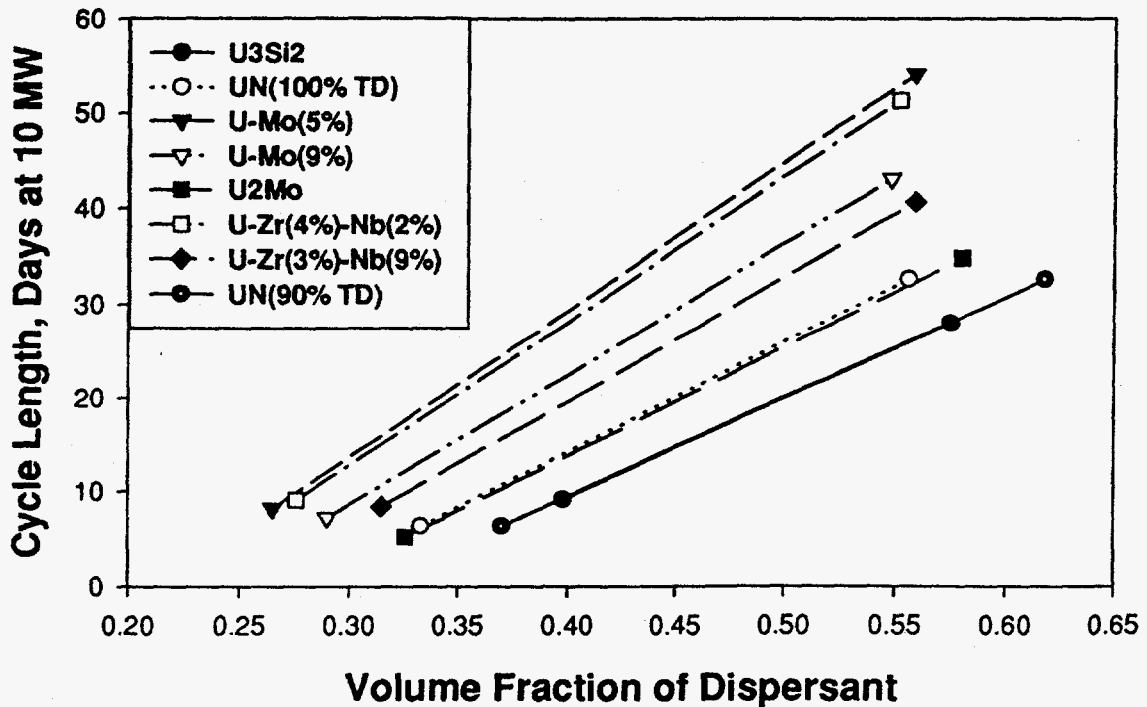

Figure 4. Cycle Length vs Dispersant Volume Fraction for Advanced Fuels in 3x3 Core

Table 5. BOEC Midplane Neutron Fluxes
(P=10.0 MW)

Fuel	VF^D %	ρ_U g/cm^3	FPD's	Region	Neutron Flux, E+14 n/cm^2-s			
					Gp 1*	Gp 2*	Gp 3*	Gp 4*
Compounds:								
U$_3$Si$_2$	42.4	4.80	12	Be Hole	1.422	1.849	1.999	3.531
				D$_2$O	0.532	1.010	1.209	3.144
U$_3$Si$_2$	50.0	5.65	20	Be Hole	1.401	1.827	1.992	3.261
				D$_2$O	0.523	0.994	1.186	2.996
UN	50.0	6.75	26	Be Hole	1.385	1.810	1.966	3.043
				D$_2$O	0.517	0.984	1.168	2.832
U$_2$Mo	50.0	6.90	25.6	Be Hole	1.382	1.814	1.958	3.012
				D$_2$O	0.517	0.984	1.168	2.807
Alloys:								
U-Zr(3%)-Nb(9%)	50.0	7.13	33	Be Hole	1.376	1.806	1.950	3.009
				D$_2$O	0.511	0.977	1.159	2.802
U-Zr(4%)-Nb(2%)	50.0	8.13	43	Be Hole	1.365	1.792	1.935	2.862
				D$_2$O	0.505	0.967	1.144	2.704
U-Mo(9%)	42.4	6.57	26	Be Hole	1.388	1.817	1.962	3.094
				D$_2$O	0.517	0.986	1.172	2.861
U-Mo(9%)	50.0	7.74	36	Be Hole	1.371	1.798	1.941	2.894
				D$_2$O	0.511	0.975	1.153	2.731
U-Mo(9%)	57.5	8.90	46	Be Hole	1.358	1.785	1.926	2.740
				D$_2$O	0.506	0.968	1.139	2.629
U-Mo(5%)	50.0	8.50	45	Be Hole	1.363	1.788	1.927	2.808
				D$_2$O	0.506	0.968	1.143	2.670

* The group boundaries (eV) are: 1.0 E+7, 8.21 E+5, 5.53 E+3, 6.25 E-1, 1.0 E-5
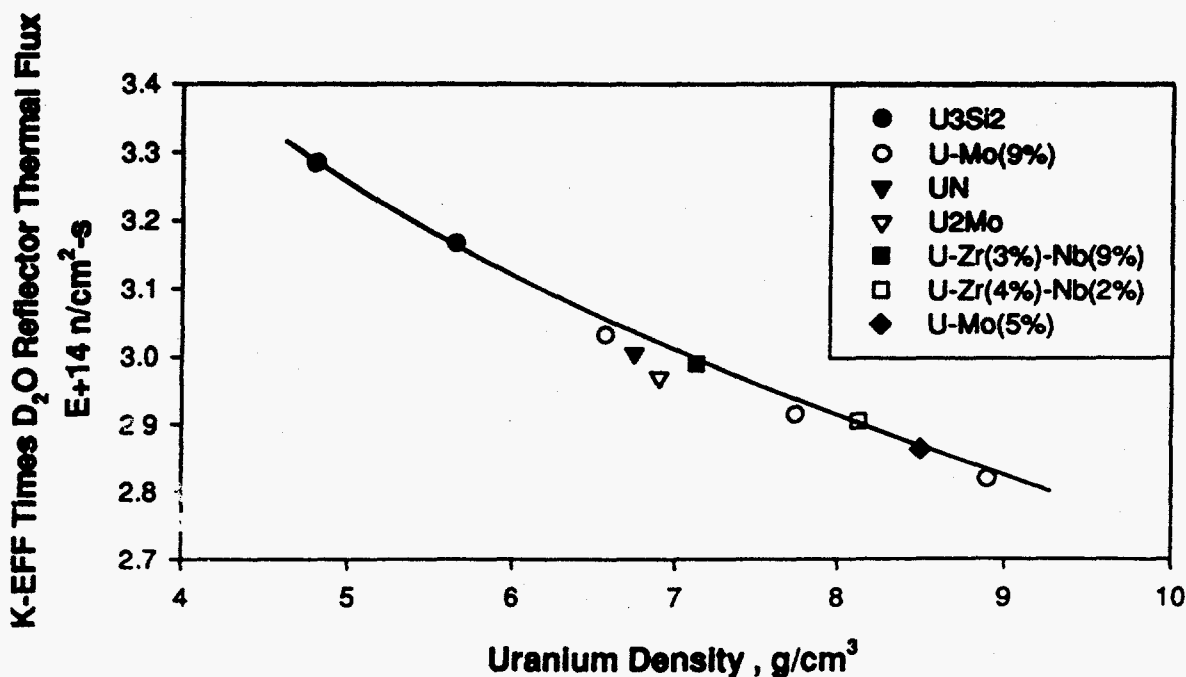
Figure 5. BOEC Thermal Neutron Flux in D$_2$O Reflector Region vs Uranium Density

If irradiation tests show that UN-Al is a successful MTR-type fuel, it could be used as a substitute for U_3Si_2-Al. Some international users of silicide have indicated particular interest in this fuel because reprocessing silicide-free fuel is simpler. For a reactor configuration similar to that described above but for a 5x5 core, calculations have shown that an LEU UN dispersion fuel with ρ_U=5.04 g/cm^3 (VF^D=37.4%) has the same cycle length as the U_3Si_2 fuel with ρ_U=4.80 g/cm^3 (VF^D=42.4%).

SUMMARY AND CONCLUSIONS

Results from this study show that dispersion fuels with certain uranium alloys offer the potential for MTR-type fuels with ρ_U > 8 gU/cm^3 if irradiation tests are successful and if requirements for licensing and final disposal are satisfied. Alloys need be chosen which stabilize the uranium, minimize parasitic neutron absorption and maximize the uranium density. From a neutronic point of view, U-Mo and U-Zr-Nb alloys are strong candidates for development. To minimize cycle length losses from neutron absorption in Mo (mostly in ^{95}Mo) and to maximize ρ_U in the U-Zr-Nb alloys it is desirable to keep the concentrations of Mo and Nb below 9 wt%. For a dispersant volume fraction of 50%, the fuels analyzed in this study are ranked by cycle length in Table 6. The correlations shown in Figs. 1 and 2 can be used to determine cycle lengths and fuel ranking for other uranium dispersants not analyzed in this study.

The maximum fabricable volume fractions for uranium alloy fuels may be above that for the brittle uranium compounds U_3Si_2 (53%, Ref. 3) and UN(52%, Ref. 4), but this awaits experimental verification. High-density dispersion fuels, with an appropriate burnable poison, could also be used to substantially increase the lifetime of fuel elements in more-conventional research reactors and thereby reduce annual disposal costs.

Table 6. Dispersion Fuels Ranked by Cycle Length for VF^D = 50%

Fuel	ρ_U-g/cm3	CL$_p$-FPD's	Relative Cycle Length
U-Mo(5%)	8.50	44.6	1.00
U-Zr(4%)-Nb(2%)	8.13	43.1	0.97
U-Mo(9%)	7.74	36.1	0.81
U-Zr(3%)-Nb(9%)	7.13	33.0	0.74
UN (100% TD)	6.75	26.5	0.59
U_2Mo	6.90	25.6	0.57
UN (90% TD)	6.08	20.1	0.45
U_3Si_2	5.65	20.1	0.45

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