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Estimated Critical Conditions for UO_2F_2 ! H_2O Systems in Fully Water-Reflected Spherical Geometry

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ESTIMATED CRITICAL CONDITIONS FOR UO_2F_2–H_2O SYSTEMS IN FULLY WATER-REFLECTED SPHERICAL GEOMETRY

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TABLE OF CONTENTS

Page

LIST OF FIGURES

LIST OF TABLES

ABSTRACT

The purpose of this report is to document reference calculations performed using the SCALE-4.0 code system to determine the critical parameters of UO_2F_2–H_2O spheres. The calculations are an extension of those documented in ORNL/CSD/TM-284. Specifically, the data for low-enriched UO_2F_2–H_2O spheres have been extended to highly enriched uranium. These calculations, together with those reported in ORNL/CSD/TM-284, provide a consistent set of critical parameters (k_4, volume, mass, mass of water) for UO_2F_2 and water over the full range of enrichment and moderation ratio.

INTRODUCTION

The purpose of this report is to document reference calculations performed to determine the critical parameters of UO_2F_2–H_2O spheres. The calculations are an extension of those documented in ORNL/CSD/TM-284.[1] Specifically, the data for low-enriched UO_2F_2–H_2O spheres have been extended to highly enriched uranium. The calculations were originally performed to supplement available data for enriched uranium. It was of interest to obtain a consistent set of critical parameters (k_4, volume, mass, mass of water) for UO_2F_2 and water over the full range of enrichment and moderation ratio.

The calculations were performed using a version of SCALE-4.0 (ref. 2) on the CRAY computer located at the Oak Ridge Gaseous Diffusion Plant in Oak Ridge, Tennessee. This computer was chosen because of the availability of free computer time. This version of SCALE is substantially the same as the configuration control version of SCALE-4.0 but is not under configuration control. The version of the code used in this analysis is not considered a QA version and has not been validated.

CALCULATIONAL METHODOLOGY AND RESULTS

The calculational methodology falls into three logical steps. The first step is a determination of the physical properties of uniform, homogeneous UO_2F_2–H_2O mixtures (density of uranium, specific gravity of the mixture, etc.) as a function of moderation ratio. The determination must be sufficiently accurate to be of value and must be valid over a wide range of moderation ratios. Because the interest is related to criticality safety, it is desirable that any inaccuracy be toward the conservative; in this case, the estimated density at a given moderation level is at or slightly greater than that which could physically occur. The second step is the calculation of the infinite media multiplication factor, k_4, for the range of enrichment and moderation ratio. The infinite media multiplication yields important information about the system: (1) it indicates where criticality is possible, (2) the moderation region where peak reactivity occurs and the reactivity characteristics of undermoderated and overmoderated systems, and (3) it is independent of the density of the system, being related only to enrichment and moderation ratio. The third step is the determination of the critical parameters over the range of interest. In this case, it was desired that the parameters be conservative with respect to geometry and reflection conditions. The calculations were performed considering full water reflection and spherical geometry. No vessel was modeled.

The density of UO_2F_2–H_2O systems as a function of the hydrogen-to-fissile (H/X) atomic ratio was calculated using a semi-empirical formulation based on the hydrogen-to-uranium (H/U) atomic ratio. This formulation appears in Appendix A of ref. 1 and has been included in Appendix A of this document for reference purposes. The formulation is discontinuous, for physical reasons, at an H/U = 4. At moderation ratios above an H/U = 4, the density formulation has a volume-additive functional form. The use of the theoretical density of uranyl fluoride dihydrate ($UO_2F_2 \cdot 2H_2O$) in a volume additive formulation, derived by considering UO_2F_2 and H_2O as a mixture (rather than a solution), yields density estimates that are in good agreement with measured densities for systems with an H/U > 4. At moderation ratios below an H/U = 4, the density is estimated using a linear fit with anhydrous UO_2F_2 at an H/U = 0 as one end point, and a density value at an H/U = 4, which bounds the densities of the various hydrates of UO_2F_2. The estimated densities used in this calculational study are also given in Appendix A. Figure A.1 depicts the uranium density as a function of the H/U moderating ratio. The scatter in the curves around an H/U = 4 is due to the use of a smooth fit through data that are discontinuous.

Parametric calculations were performed over the range of uranium enrichments from 1.4 to 100 wt % ^{235}U and over a broad range of H/X ratios. The infinite media multiplication for UO_2F_2–H_2O systems was estimated using the CSAS1X[3] control module of the CSAS4 control sequence in SCALE-4.0 and the SCALE 27-group cross-section library.[4] Critical radius searches were performed using XSDRNPM-S[5] with cross-section preprocessing performed with the CSASI control module of the CSAS4 control sequence. The critical radii of water-reflected UO_2F_2–H_2O spheres were estimated. From these results, estimates of the critical volume, critical mass of uranium, and mass of water for criticality were made. These results are presented graphically in Figs. 1–5 and tabulated in Table B.1. In the figures, the dashed curves represent an extrapolation or significant interpolation of the calculated data.

Figures 1 through 5 are presented as a function of the H/X moderating ratio. Neutronic characteristics are more clearly depicted as a function of H/X as opposed to H/U. (The H/X and H/U moderating ratios are interrelated by enrichment. This relationship may be approximated as H/X • H/U ÷ E.) One neutronic characteristic is that optimum moderation for minimum ^{235}U critical mass in homogeneous water-moderated uranium systems occurs at an H/X of approximately 500 independent of enrichment. Another useful characteristic is that optimum moderation for minimum volume systems usually occurs in the region of peak k_4.

Selected iso-H/U values are shown in several of the figures. The physical characteristics of UO_2F_2–H_2O systems are related to the H/U of the system. An H/U of 4 or less represents a hydrated solid compound or a mixture of hydrated solids. An H/U of about 16 is the saturation limit for UO_2F_2 and water. At values below an H/U = 16, UO_2F_2 and H_2O are no longer a solution but a mixture or slurry of hydrated solid UO_2F_2AH_2O at some bulk density with intermixed water. At H/U ratios greater than about 16, UO_2F_2 and water form a solution.

Figure 6 is a hybrid curve showing the relationship between the mass of water for criticality as a function of the critical mass of ^{235}U. For a given enrichment, points that fall below the enrichment curve and to the left are subcritical due to insufficient water or insufficient mass of uranium for the water present. The curve indicates that enrichments of 10% and greater may be made critical in the absence of a moderator, although the mass may be substantial. UO_2F_2 and water systems at enrichments below about 7% cannot be made critical in the absence of a moderator. It appears that a minimum of 12 kg of water is required for criticality of UO_2F_2-H_2O enriched to less than 7%. Also note that uranium enriched to less than about 2% cannot be made critical as a solution (H/U > 16) but only as a mixture or slurry of UO_2F_2A2H_2O and H_2O.

Figure 7 shows the critical mass of uranium versus the critical volume of the system. The effect of the discontinuity in the density relationship on the predicted critical mass and volume is clearly seen in the irregular shape of the curve for 50% enrichment around an H/X = 10.

Figure 8 shows the trend of peak k_4 as a function of enrichment, along with k_4 for unmoderated UO_2F_2. Extrapolation of the peak k_4 curve indicates a subcritical enrichment of about 1 wt % ^{235}U. Interpolation of the unmoderated k_4 curve indicates that UO_2F_2 enriched to less than about 9 wt % ^{235}U cannot be made critical in the complete absence of a moderator.

DISCUSSION

The data are presented for information purposes and should be used with caution. No safety factors are incorporated into the reported results. The estimates are based on critical water-reflected spherical geometry. Little independent review or checking has been performed. It is recommended that if these data are to be used in any safety evaluation, they be substantially checked and verified against calculations performed with a validated version of SCALE and/or other accepted criticality codes and cross sections.

Certain characteristics of the SCALE 27-group ENDF/B-IV library may bias the calculations. These uncertainties should be taken into account in using the data presented in Figs. 1–6. The calculated k_4 may be as much as 1% low for homogeneous low-enriched systems. This is due, in part, to inadequacies in the ENDF/B-IV ^{238}U resonance cross sections. Also, the unresolved resonance region for ^{238}U was processed at a fixed F_p = 50,000 in the SCALE 27-group library. Both of these cause the ^{238}U capture to be too high. This bias is a function of the enrichment and moderation level (neutron energy spectrum) of the system.

Calculated k_4 for highly enriched uranium may be biased as much as 1% high. This is due, in part, to the unresolved resonance region of ^{235}U being processed at a fixed F_p = 50,000 in the SCALE 27-group library. This bias is also a function of the enrichment and moderation level of the system. The critical systems presented are thought to be accurate to within ±1.5% in k_{eff} (0.985 # k_{eff} # 1.015) over the range of conditions calculated, assuming there is no uncertainty assigned to the density of the systems. The effect of this accuracy on the estimated critical volume, mass of uranium (total U and ^{235}U), and mass of water for criticality is a function of k_4 of the system and could be quite large for systems with low reactivity (k_4 • 1).

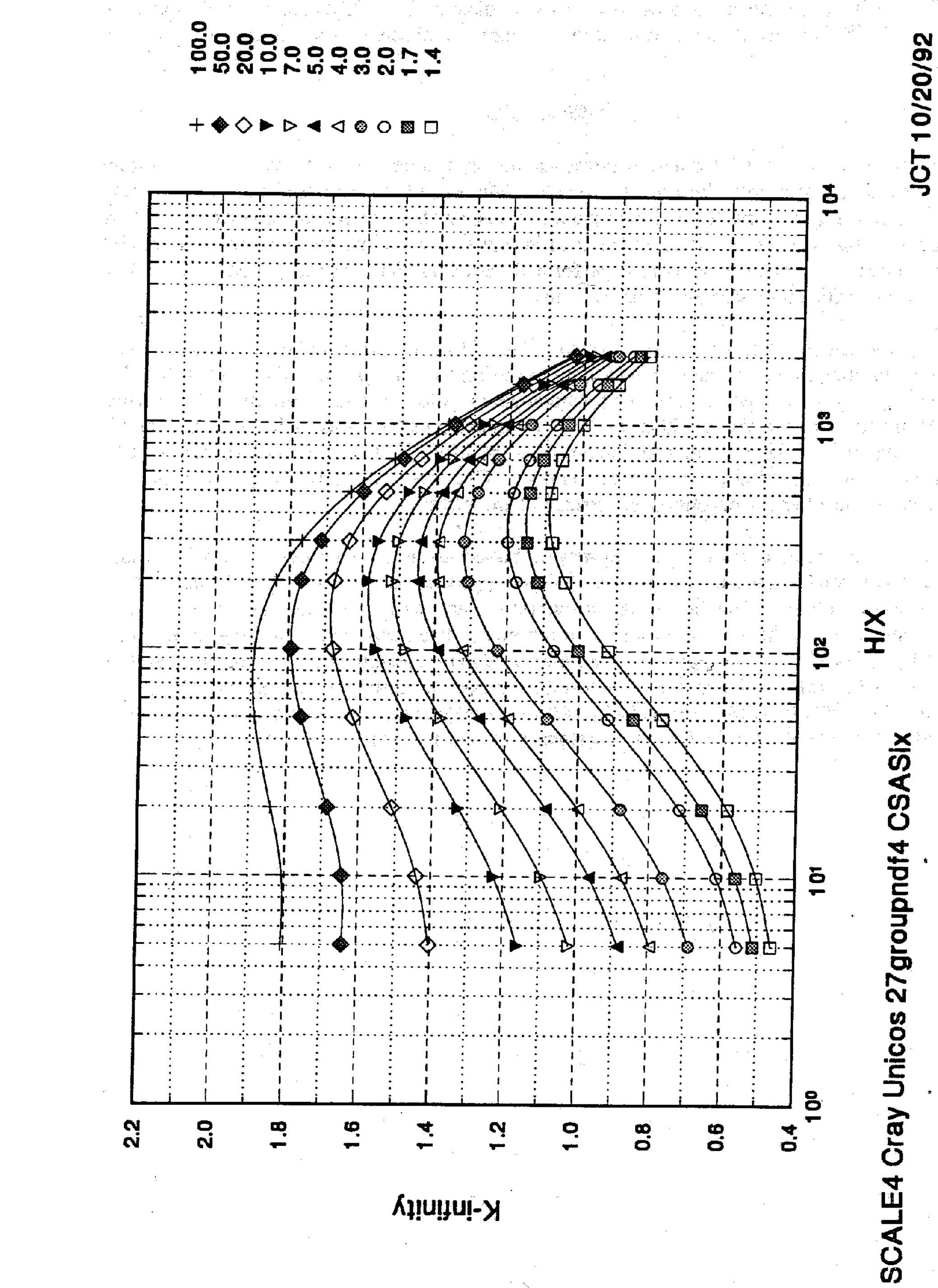

Fig. 1. k_4 vs H/X for UO_2F_2–H_2O.

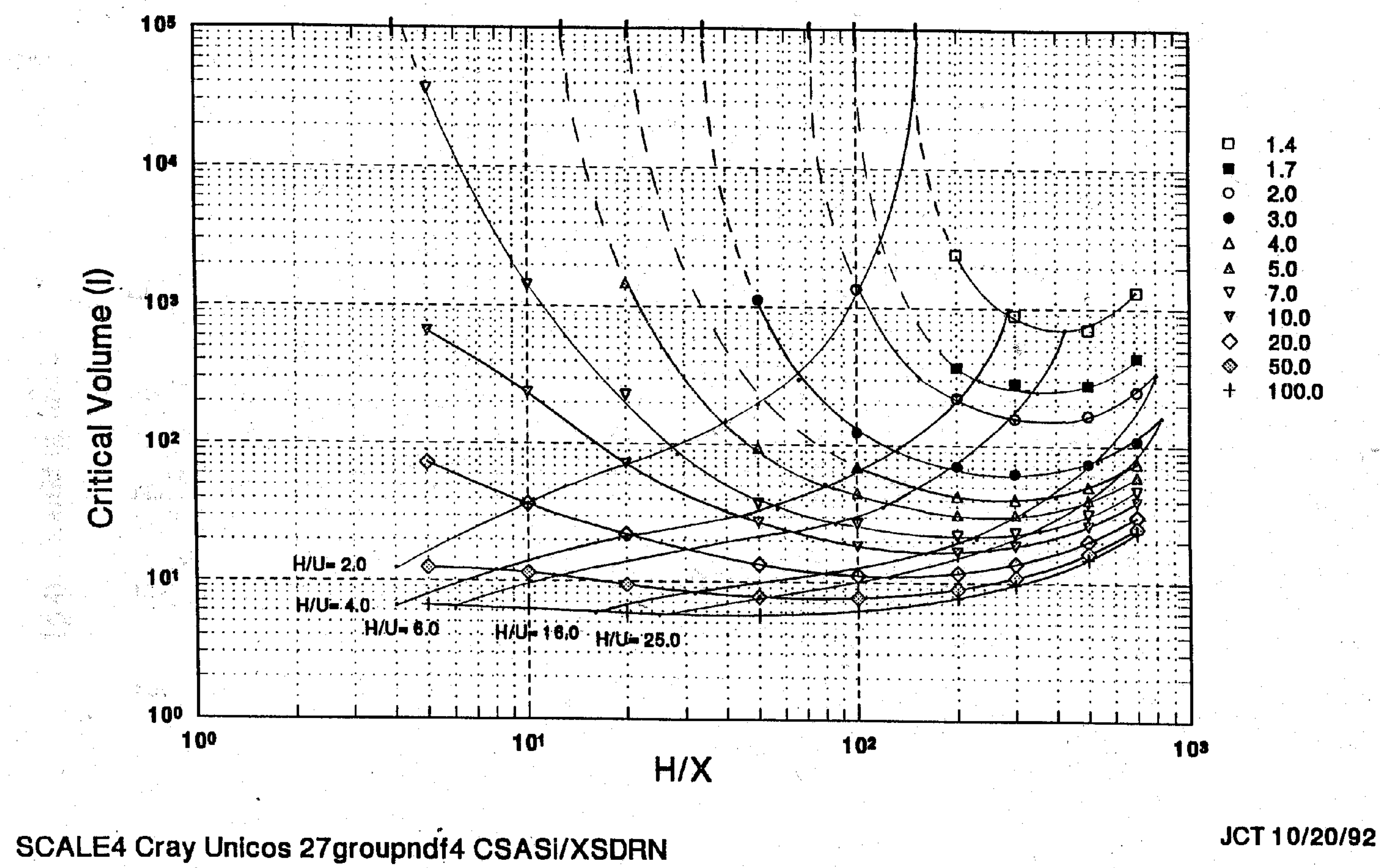

Fig. 2. Critical volume vs H/X for UO_2F_2–H_2O in spherical H_2O reflected systems.

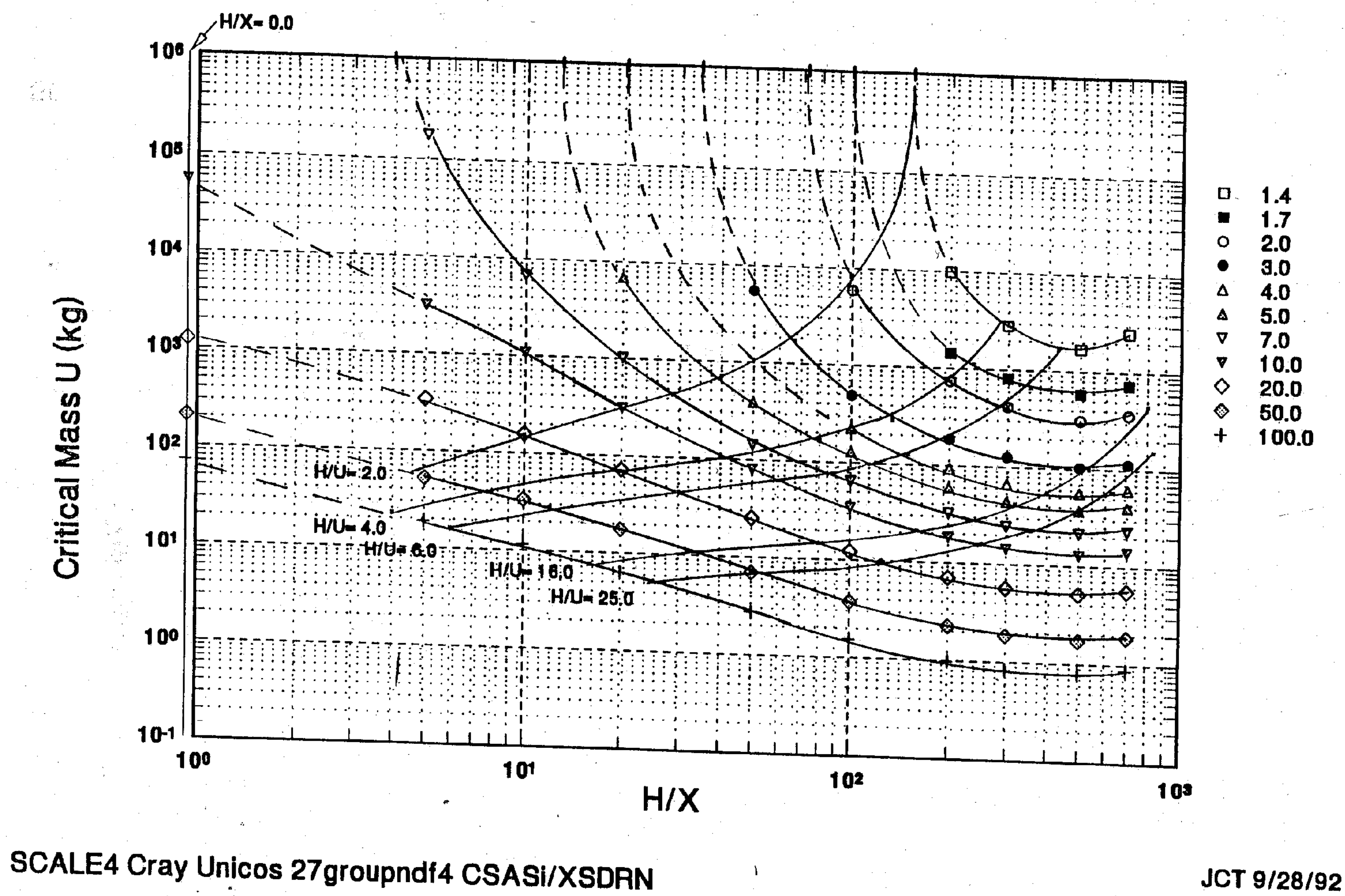

Fig. 3. Critical mass U vs H/X for UO_2F_2–H_2O in spherical H_2O-reflected systems.

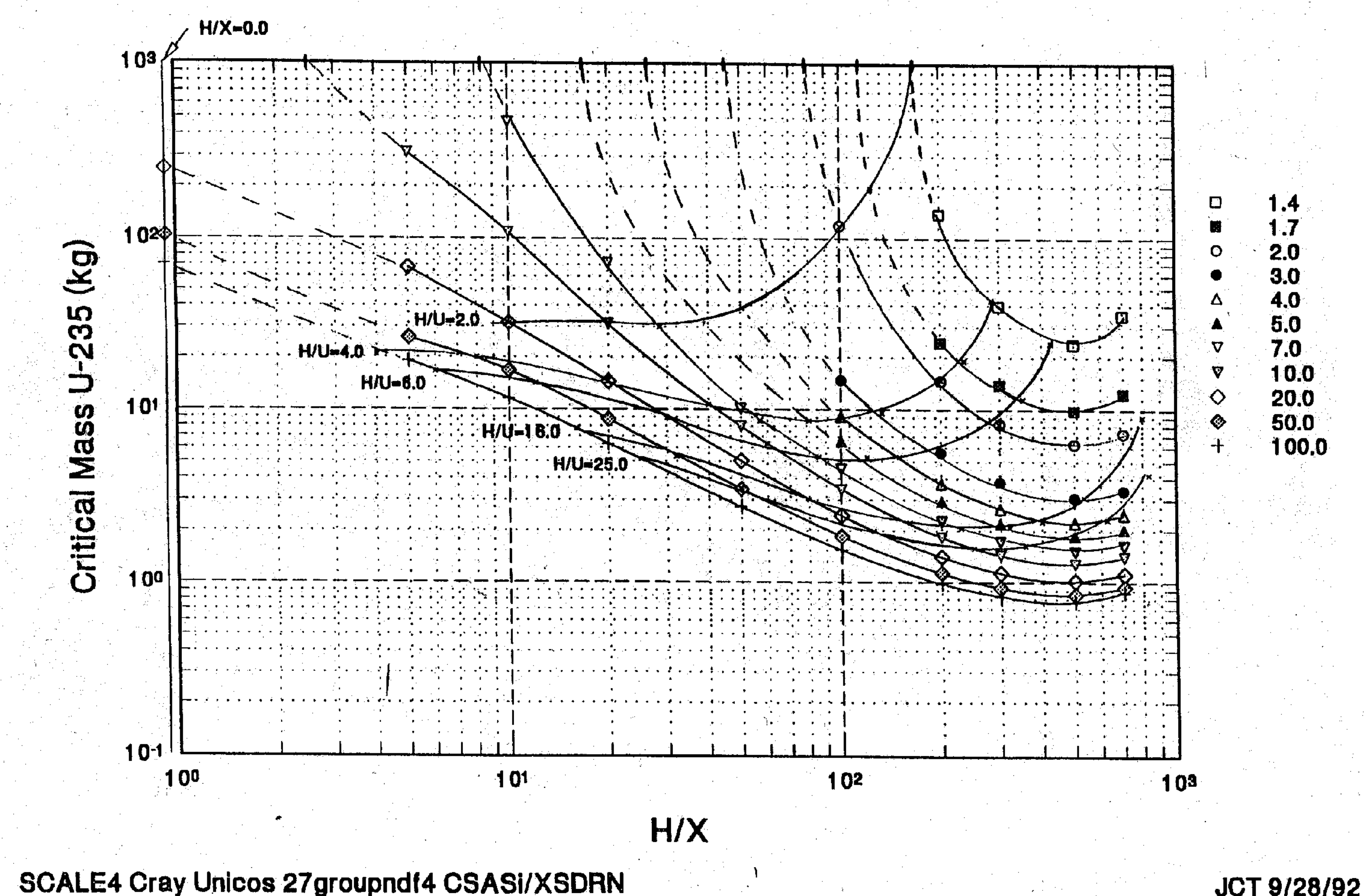

Fig. 4. Critical mass ^{235}U vs H/X for UO_2F_2–H_2O in spherical H_2O-reflected systems.

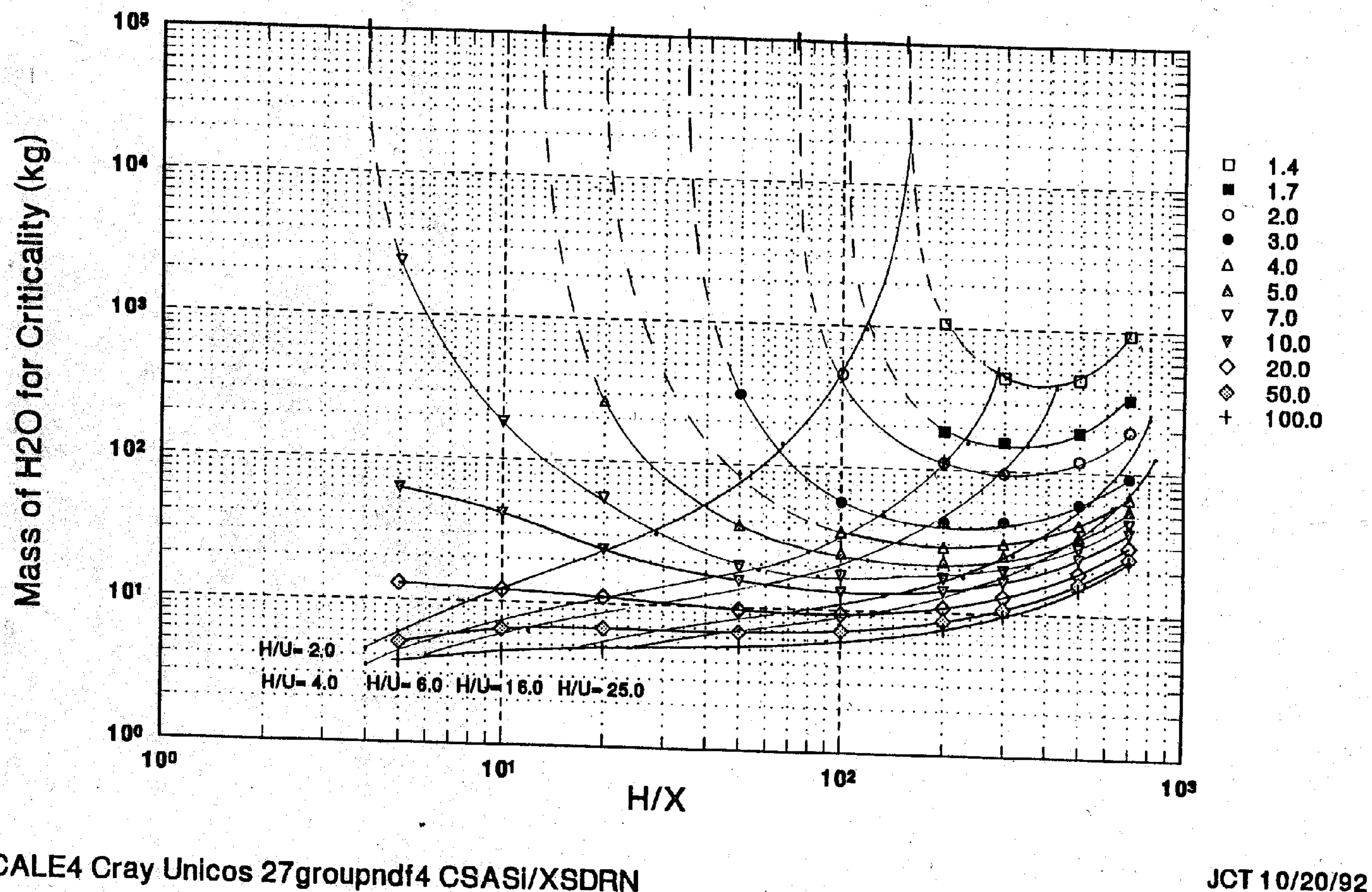

Fig. 5. Mass of H_2O for criticality vs H/X for UO_2F_2–H_2O in spherical H_2O-reflected systems.

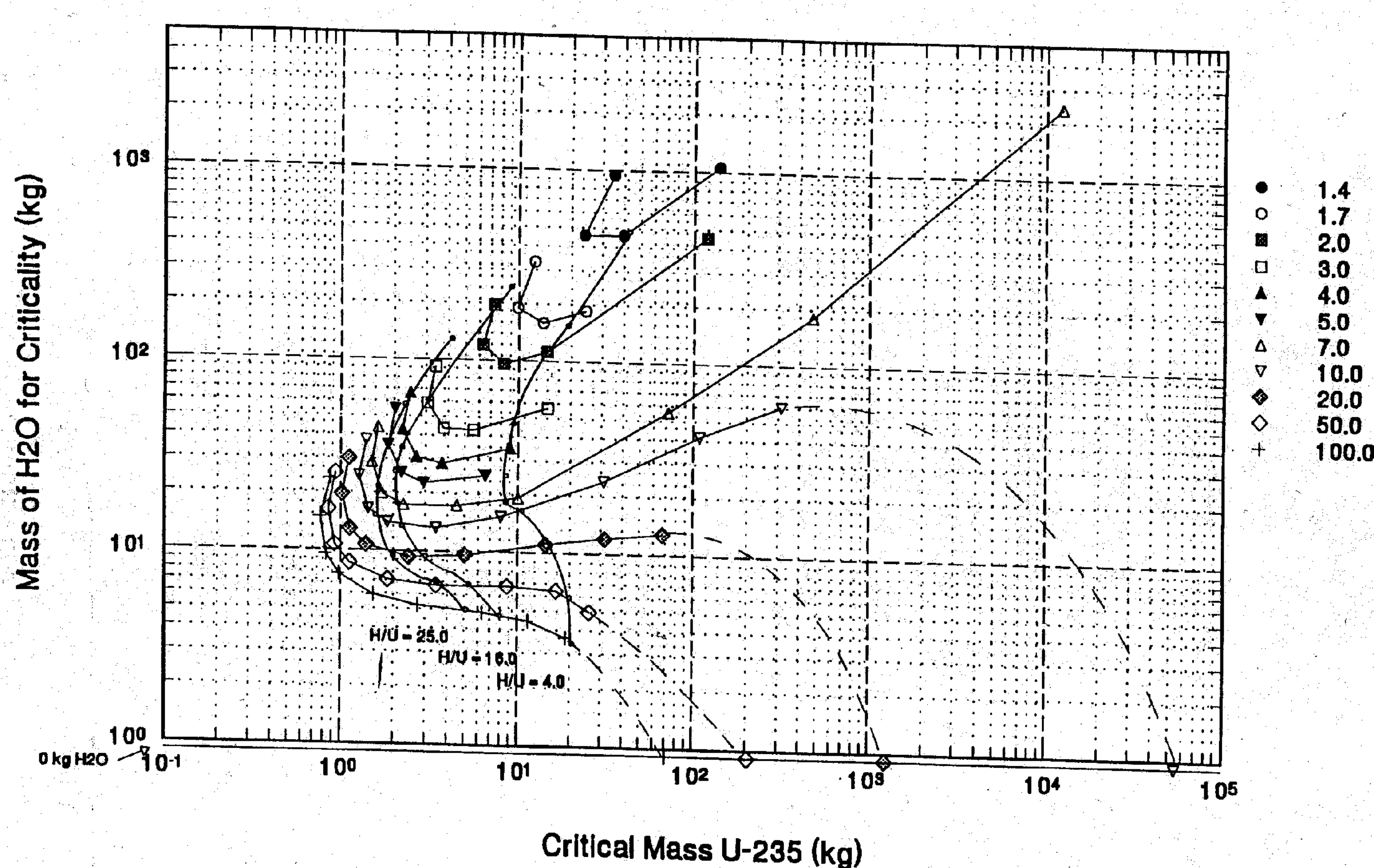

Fig. 6. Mass of H_2O for criticality vs critical mass ^{235}U in spherical H_2O-reflected systems.

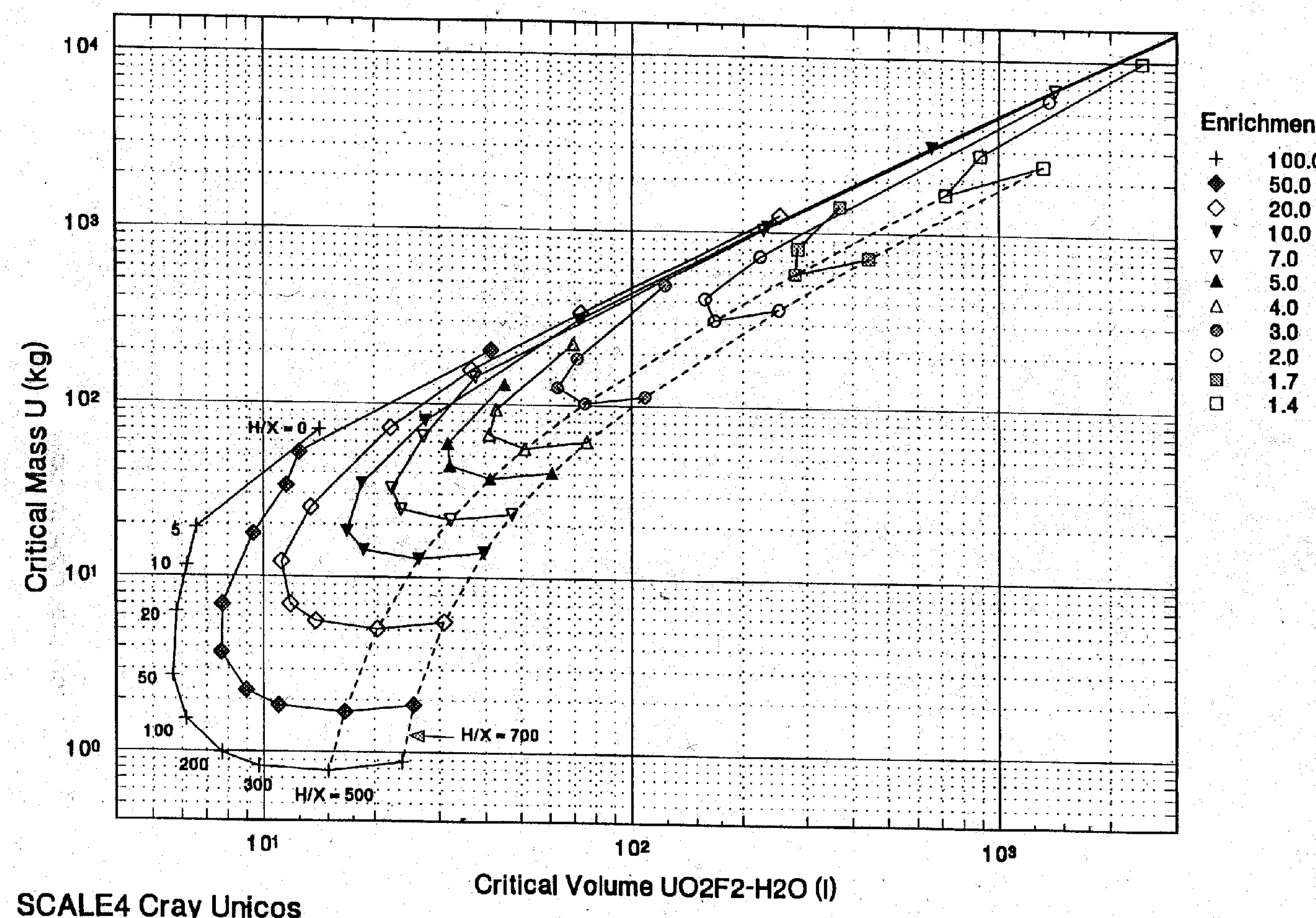

Fig. 7. Critical mass U (kg) vs critical volume (l).

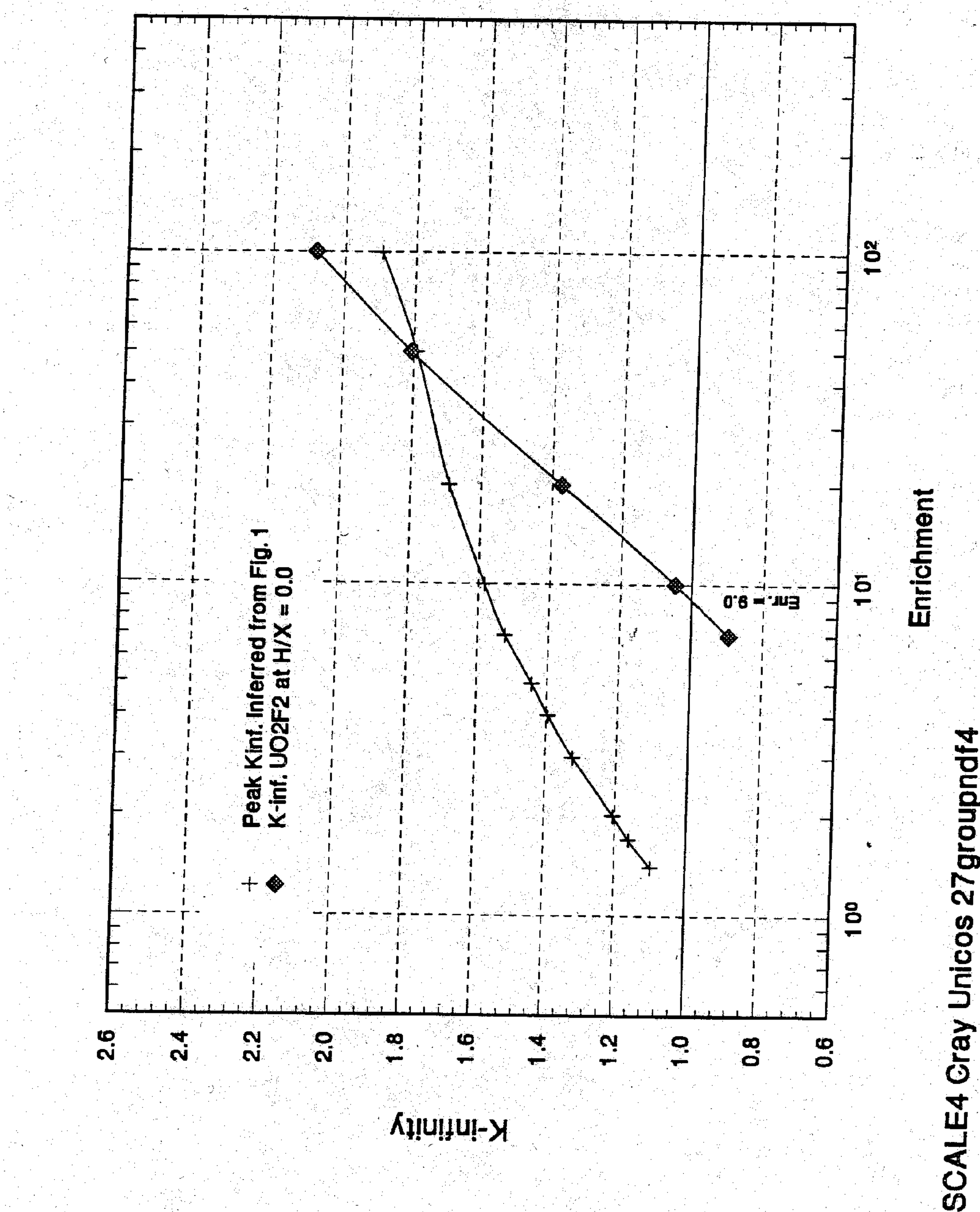

Fig. 8. k_4 vs enrichment for UO_2F_2–H_2O.

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3. N. F. Landers and L. M. Petrie, "CSAS4: An Enhanced Criticality Safety Analysis Module with an Optimum Pitch Search Option," Sect. C4 of *SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation*, NUREG/CR-0200, Rev. 4 (ORNL/NUREG/CSD-2/R4), Vols. I, II, and III (draft February 1990), Martin Marietta Energy Systems, Inc., Oak Ridge Natl. Lab. Available from Radiation Shielding Information Center as CCC-545.

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APPENDIX A

DENSITY RELATIONSHIPS

The purpose of this Appendix is to present relationships for uranium density as a function of H/U ratio for uranium compounds. Water and HF have been considered as the principal moderators. The density of homogeneous mixtures may be estimated based on the principle of additive molar volume. One form of this is given in Eq. (A.1), where the uranium density in a mixture is expressed as a function of the H/U:

$$\rho_u = \frac{M_u}{\frac{V_{uc}}{N} + \left(\frac{H}{U} - M * Y\right) * \frac{V_m}{M}} \, , \qquad \text{(A.1)}$$

where

D_u	=	uranium density,
M_u	=	molecular weight of uranium at a given enrichment,
V_{uc}	=	molar volume of the uranium compound (M_{uc}/D_{uc}),
N	=	number of uranium atoms in the uranium compound,
$\frac{V_{uc}}{N}$	=	specific molar volume of uranium compound,
V_m	=	molar volume of the moderating compound (M_m/D_m),
M	=	number of hydrogen atoms in the moderating compound,
$\frac{V_m}{M}$	=	specific molar volume of hydrogenous compound,
$\frac{H}{U}$	=	the H/U moderating ratio,
Y	=	number of hydrated moderating molecules in uranium compound (hydrated water for water-moderated systems, hydrated HF for HF-moderated systems).

In Eq. (A.1), M is used for molecular weight, D is used for density, and V is used for molar volume ($V = M/D$). The subscripts u, uc, and m refer to uranium, uranium compound, and moderating compound, respectively. The form of Eq. (A.1) is general and requires a minimum of tabulated data to represent a large number of systems. The "specific molar volume" (V_{uc}/N and V_m/M) used in Eq. (A.1) is a contrived quantity which is defined as the molar volume (the molecular weight of a compound divided by its density) divided by the number of atoms of the nuclide of interest in the compound, the number of uranium atoms for the compound and the number of hydrogen atoms for the hydrogenous moderator. Equation (A.1) could be simplified, but, in doing so, the physical significance of the quantities that are involved is lost. As it stands, Eq. (A.1) is quite general and may be easily adapted for use on more complex mixtures and solutions.

The quantity defined as the specific molar volume has the interesting property that the ratio of the molecular weight of a compound divided by the theoretical density of that compound is nearly constant, independent of the isotopic distribution of the elements in the molecule. For example, the specific molar volume for the uranium compound is independent of the uranium enrichment in the compound. This condition exists because the theoretical density of the uranium compound has the same variation with enrichment as the molecular weight of uranium. Table A.1 lists several uranium compounds and moderating materials. The molecular weight and theoretical densities are given, along with the specific molar volumes. Variations of the theoretical density of the mixtures due to temperature may be taken into account by using the appropriate theoretical density of the compound in computing the specific molar volume. The values listed in Table A.1 are based on a temperature of 23°C. The density variation as a function of temperature is given in Table A.2 for several of the compounds.

The use of Eq. (A.1), with the specific molar volumes given in Table A.1, should yield maximum uranium densities for a given H/U and are therefore conservative. (Note that the molecular weights given in Table A.1 are not consistent with those in SCALE-4.0. The molecular weights in Table A.1 are those weights that were used in the original testing of the density relationships. Revising the molecular weights to those in SCALE-4.0 would not significantly affect the specific molar volumes tabulated; however, revisions were not made here in order to maintain traceability to my notes.)

Uranyl fluoride and uranyl nitrate form solutions when moderated by water. They also form water hydrates at low H/U ratios. This complicates the theoretical predictions of the uranium density. Because of these characteristics, the uranyl fluoride and uranyl nitrate systems have been treated in more detail.

E. J. Barber suggested that the relationship for uranyl fluoride be discontinuous at H/U = 4 in order to account for the various hydrates that exist. Only mixtures of hydrated salts exist at H/U < 4. Given this consideration, the density of uranyl fluoride may be estimated by Eqs. (A.2) and (A.3), where Eq. (A.3) is the specific form of Eq. (A.1).

URANYL FLUORIDE

H/U < 4

$$\rho_u = 4.96 - 0.32 * \left(\frac{H}{U}\right) . \tag{A.2}$$

H/U $ 4

$$\rho_u = \frac{M_u}{V_{UO_2F_2 * 2H_2O} + \left(\frac{H}{U} - 4\right) * \frac{V_{H_2O}}{2}} . \tag{A.3}$$

Table A.1. Properties of uranium and moderating compounds

Compound	Molecular weight (M°)	Wt % U in compound	Density (g/cc) @23°C	Specific molar volume (V_{uc}/N) or (V_m/M)
U	238.029	100.00	19.05	12.4950
UO_2	270.028	88.15	10.96	24.6376
U_3O_8	842.082	84.80	8.30	33.8187
UF_6(solid)	352.019	67.62	5.075[a]	69.3633
UO_2F_2–$2H_2O$	344.057	69.18	4.76	72.2809
$UO_2(NO_3)_2$A$6H_2O$	502.134	47.40	2.745[a]	182.927
H_2O	18.016	---	0.9977[a]	9.0287
HF(liquid)	20.0063	---	0.9516[a]	21.0239
HNO_3	63.0128	---	1.9447	32.4023

[a] The density is a function of temperature, as given in Table 2. The tabulated volume is at 23°C.

Table A.2. Temperature variation of several compounds

Compound	Density @ t °C (g/cc)	Notes[a]
UF_6(solid)	5.194 – 0.005168*t	(1)
UF_6(liquid)	1.670 + 0.15203*(230.2 – t)$^{1/2}$	(2)
$UO_2(NO_3)_2$A$6H_2O$	2.745*(1 – (t – 23.)*0.0015)	(3)
H_2O	0.99987/[1. – 6.427-5*t + 8.5053-6*t^2 – 6.79-8*t^3]	(4)
HF(liquid)	1.0020 – 0.0022625*t + 3.125-6*t^2	(5)
HF(solid)	1.562 – 0.000979*t	(5)
HNO_3	1.95 – (t – 20.)*0.00176	(3)

[a] Sources of values:

(1) E. J. Barber, *Estimation of Pressures Developed by Retrained Solid UF_6*, K/ET-307, Union Carbide Corp., Nucl. Div., Oak Ridge Gaseous Diffusion Plant, 1979.

(2) E. J. Barber, *Relationship of Pressure to Temperature Rise in Overfilled Cylinders*, K/ET-194, Union Carbide Corp., Nucl. Div., Oak Ridge Gaseous Diffusion Plant, (1979).

(3) Fit to measured data by W. C. Jordan, ORNL, 1988.

(4) Fit to specific volume data for water 15 < t < 30°C.

(5) I. G. Ryss, *Chemistry of Fluorine and Its Inorganic Compounds*, AEC-tr-3927 Part 1, Chapter 3 - Hydrogen Flouride. Data for solid - p. 65 (LeBoucher, Fischer, Biltz). Data for liquid -p. 66 (Simons and Bouknight).

URANYL NITRATE

$$\rho_u = \frac{M_u * \left[1 + \frac{M^+}{1000} * \left(V_{H_2O} - V_{HNO_3}\right)\right]}{V_{UNH} + \left(\frac{H}{U} - 12\right) * \frac{V_{H_2O}}{2}} \quad . \qquad \text{(A.4)}$$

It was found that, for uranyl nitrate, the use of theoretical densities in Eq. (A.1) did not yield accurate uranium densities over the full range of H/U. In addition, the inclusion of the excess acid into Eq. (A.1) and the use of theoretical densities for the excess acid gave unreasonable density effects. The molar volume of uranyl nitrate hexahydrate and nitric acid given in Table A.1 were adjusted for a best fit to data on uranyl nitrate solutions with excess acid using Eq. (A.4). Equation (A.4) is the specific form of Eq. (A.1) for uranyl nitrate, including the excess acid. Equation (A.4) is similar to that in ARH-600, but it is believed to be a better estimate for the nitric acid system.

Mixtures of various hydrate forms must be assumed for H/U ratios <12 in uranyl nitrate systems. Only limited information was found for these systems. It is believed that the formulations in Eq. (A.4) may significantly overpredict uranium densities at H/U < 12.

Table A.3 gives the theoretical density for UO_2F_2–H_2O mixtures as predicted by Eqs. (A.2) and (A.3) for a temperature of 20°C. These values were used in the calculations presented in the previous section entitled "Calculational Methodology and Results." The uranium density, uranium compound density, and moderator densities are tabulated for the specified H/X (and corresponding H/U) moderation ratios. Uranium enrichments of 100, 50, 20, 10, 7, 5, 4, 3, 2, 1.7, and 1.4% ^{235}U were considered. The predicted uranium densities are plotted as a function of the H/U and H/X moderation ratios in Figs. A.1 and A.2, respectively.

Table A.3. Physical properties of UO_2F_2–H_2O mixtures

		UO_2F_2–H_20		
H/X	H/U	U den.	UO_2F_2 den.	H_2O den.
1.0E+02 % Enriched				
5.000E+00	5.000E+00	2.8911E+00	3.7519E+00	5.5394E-01
1.000E+01	9.999E+00	1.8594E+00	2.4130E+00	7.1252E-01
2.000E+01	2.000E+01	1.0850E+00	1.4080E+00	8.3154E-01
5.000E+01	5.000E+01	4.8233E-01	6.2595E-01	9.2417E-01
1.000E+02	9.999E+01	2.5046E-01	3.2504E-01	9.5980E-01
2.000E+02	2.000E+02	1.2769E-01	1.6572E-01	9.7867E-01
3.000E+02	3.000E+02	8.5691E-02	1.1121E-01	9.8513E-01
5.000E+02	5.000E+02	5.1687E-02	6.7078E-02	9.9035E-01
7.000E+02	6.999E+02	3.7004E-02	4.8022E-02	9.9261E-01
1.000E+03	9.999E+02	2.5947E-02	3.3673E-02	9.9431E-01
1.500E+03	1.500E+03	1.7321E-02	2.2479E-02	9.9564E-01
2.000E+03	2.000E+03	1.2999E-02	1.6870E-02	9.9630E-01
5.0E+01 % Enriched				
5.000E+00	2.516E+00	4.1549E+00	5.3843E+00	3.9809E-01
1.000E+01	5.032E+00	2.8991E+00	3.7569E+00	5.5553E-01
2.000E+01	1.006E+01	1.8626E+00	2.4137E+00	7.1383E-01
5.000E+01	2.516E+01	8.9868E-01	1.1646E+00	8.6105E-01
1.000E+02	5.032E+01	4.8251E-01	6.2528E-01	9.2461E-01
2.000E+02	1.006E+02	2.5050E-01	3.2462E-01	9.6004E-01
3.000E+02	1.510E+02	1.6916E-01	2.1921E-01	9.7246E-01
5.000E+02	2.516E+02	1.0256E-01	1.3290E-01	9.8264E-01
7.000E+02	3.522E+02	7.3586E-02	9.5359E-02	9.8706E-01
1.000E+03	5.032E+02	5.1685E-02	6.6978E-02	9.9041E-01
1.500E+03	7.548E+02	3.4548E-02	4.4770E-02	9.9302E-01
2.000E+03	1.006E+03	2.5945E-02	3.3622E-02	9.9434E-01
2.0E+01 % Enriched				
5.000E+00	1.010E+00	4.6367E+00	6.0034E+00	1.7770E-01
1.000E+01	2.020E+00	4.3135E+00	5.5849E+00	3.3062E-01
2.000E+01	4.041E+00	3.2684E+00	4.2317E+00	5.0103E-01
5.000E+01	1.010E+01	1.8646E+00	2.4142E+00	7.1461E-01
1.000E+02	2.020E+01	1.0867E+00	1.4070E+00	8.3296E-01
2.000E+02	4.041E+01	5.9242E-01	7.6703E-01	9.0817E-01
3.000E+02	6.061E+01	4.0720E-01	5.2722E-01	9.3635E-01
5.000E+02	1.010E+02	2.5054E-01	3.2439E-01	9.6018E-01
7.000E+02	1.414E+02	1.8093E-01	2.3426E-01	9.7077E-01
1.000E+03	2.020E+02	1.2771E-01	1.6535E-01	9.7887E-01
1.500E+03	3.031E+02	8.5694E-02	1.1095E-01	9.8526E-01
2.000E+03	4.041E+02	6.4481E-02	8.3487E-02	9.8849E-01

Table A.3. (continued)

		UO_2F_2–H_20		
H/X	H/U	U den.	UO_2F_2 den.	H_2O den.
1.0E+01 % Enriched				
5.000E+00	5.057E-01	4.7982E+00	6.2106E+00	9.1944E-02
1.000E+01	1.011E+00	4.6363E+00	6.0011E+00	1.7769E-01
2.000E+01	2.023E+00	4.3126E+00	5.5822E+00	3.3056E-01
5.000E+01	5.057E+00	2.9056E+00	3.7610E+00	5.5679E-01
1.000E+02	1.011E+01	1.8653E+00	2.4144E+00	7.1487E-01
2.000E+02	2.023E+01	1.0869E+00	1.4069E+00	8.3314E-01
3.000E+02	3.034E+01	7.6692E-01	9.9269E-01	8.8176E-01
5.000E+02	5.057E+01	4.8269E-01	6.2479E-01	9.2495E-01
7.000E+02	7.080E+01	3.5217E-01	4.5585E-01	9.4479E-01
1.000E+03	1.011E+02	2.5055E-01	3.2431E-01	9.6023E-01
1.500E+03	1.517E+02	1.6918E-01	2.1899E-01	9.7259E-01
2.000E+03	2.023E+02	1.2771E-01	1.6531E-01	9.7889E-01
7.0E+00 % Enriched				
5.000E+00	3.542E-01	4.8467E+00	6.2729E+00	6.5012E-02
1.000E+01	7.083E-01	4.7333E+00	6.1262E+00	1.2698E-01
2.000E+01	1.417E+00	4.5067E+00	5.8328E+00	2.4180E-01
5.000E+01	3.542E+00	3.8267E+00	4.9527E+00	5.1330E-01
1.000E+02	7.083E+00	2.3760E+00	3.0751E+00	6.3741E-01
2.000E+02	1.417E+01	1.4501E+00	1.8768E+00	7.7804E-01
3.000E+02	2.125E+01	1.0435E+00	1.3505E+00	8.3980E-01
5.000E+02	3.542E+01	6.6854E-01	8.6526E-01	8.9675E-01
7.000E+02	4.958E+01	4.9182E-01	6.3655E-01	9.2360E-01
1.000E+03	7.083E+01	3.5218E-01	4.5581E-01	9.4481E-01
1.500E+03	1.062E+02	2.3906E-01	3.0940E-01	9.6199E-01
2.000E+03	1.417E+02	1.8094E-01	2.3418E-01	9.7082E-01
5.0E+00 % Enriched				
5.000E+00	2.532E-01	4.8790E+00	6.3143E+00	4.6776E-02
1.000E+01	5.064E-01	4.7980E+00	6.2095E+00	9.1999E-02
2.000E+01	1.013E+00	4.6359E+00	5.9997E+00	1.7778E-01
5.000E+01	2.532E+00	4.1498E+00	5.3706E+00	3.9785E-01
1.000E+02	5.064E+00	2.9054E+00	3.7601E+00	5.5710E-01
2.000E+02	1.013E+01	1.8648E+00	2.4134E+00	7.1513E-01
3.000E+02	1.519E+01	1.3730E+00	1.7769E+00	7.8980E-01
5.000E+02	2.532E+01	8.9889E-01	1.1633E+00	8.6180E-01
7.000E+02	3.545E+01	6.6817E-01	8.6474E-01	8.9683E-01
1.000E+03	5.064E+01	4.8243E-01	6.2436E-01	9.2504E-01
1.500E+03	7.596E+01	3.2968E-01	4.2667E-01	9.4824E-01
2.000E+03	1.013E+02	2.5040E-01	3.2407E-01	9.6027E-01

Table A.3 (continued)

		UO_2F_2–H_20		
H/X	H/U	U den.	UO_2F_2 den.	H_2O den.
4.0E+00 % Enriched				
5.000E+00	2.026E-01	4.8952E+00	6.3351E+00	3.7540E-02
1.000E+01	4.051E-01	4.8304E+00	6.2512E+00	7.4087E-02
2.000E+01	8.102E-01	4.7007E+00	6.0834E+00	1.4420E-01
5.000E+01	2.026E+00	4.3118E+00	5.5801E+00	3.3067E-01
1.000E+02	4.051E+00	3.2708E+00	4.2330E+00	5.0167E-01
2.000E+02	8.102E+00	2.1769E+00	2.8172E+00	6.6777E-01
3.000E+02	1.215E+01	1.6313E+00	2.1111E+00	7.5061E-01
5.000E+02	2.026E+01	1.0866E+00	1.4062E+00	8.3331E-01
7.000E+02	2.836E+01	8.1462E-01	1.0542E+00	8.7461E-01
1.000E+03	4.051E+01	5.9225E-01	7.6646E-01	9.0838E-01
1.500E+03	6.077E+01	4.0705E-01	5.2679E-01	9.3649E-01
2.000E+03	8.102E+01	3.1009E-01	4.0130E-01	9.5122E-01
3.0E+00 % Enriched				
5.000E+00	1.519E-01	4.9114E+00	6.3559E+00	2.8245E-02
1.000E+01	3.038E-01	4.8628E+00	6.2930E+00	5.5931E-02
2.000E+01	6.077E-01	4.7655E+00	6.1672E+00	1.0962E-01
5.000E+01	1.519E+00	4.4739E+00	5.7897E+00	2.5729E-01
1.000E+02	3.038E+00	3.9877E+00	5.1606E+00	4.5866E-01
2.000E+02	6.077E+00	2.6144E+00	3.3833E+00	6.0140E-01
3.000E+02	9.115E+00	2.0092E+00	2.6001E+00	6.9328E-01
5.000E+02	1.519E+01	1.3734E+00	1.7773E+00	7.8980E-01
7.000E+02	2.127E+01	1.0432E+00	1.3500E+00	8.3992E-01
1.000E+03	3.038E+01	7.6674E-01	9.9225E-01	8.8190E-01
1.500E+03	4.558E+01	5.3183E-01	6.8825E-01	9.1756E-01
2.000E+03	6.077E+01	4.0711E-01	5.2684E-01	9.3649E-01
2.0E+00 % Enriched				
5.000E+00	1.013E-01	4.9276E+00	6.3767E+00	1.8890E-02
1.000E+01	2.026E-01	4.8952E+00	6.3347E+00	3.7531E-02
2.000E+01	4.051E-01	4.8304E+00	6.2509E+00	7.4068E-02
5.000E+01	1.013E+00	4.6359E+00	5.9992E+00	1.7772E-01
1.000E+02	2.026E+00	4.3118E+00	5.5798E+00	3.3058E-01
2.000E+02	4.051E+00	3.2717E+00	4.2338E+00	5.0167E-01
3.000E+02	6.077E+00	2.6147E+00	3.3836E+00	6.0140E-01
5.000E+02	1.013E+01	1.8655E+00	2.4141E+00	7.1513E-01
7.000E+02	1.418E+01	1.4500E+00	1.8764E+00	7.7819E-01
1.000E+03	2.026E+01	1.0869E+00	1.4065E+00	8.3331E-01
1.500E+03	3.038E+01	7.6684E-01	9.9235E-01	8.8190E-01
2.000E+03	4.051E+01	5.9240E-01	7.6661E-01	9.0838E-01

Table A.3 (continued)

		UO_2F_2–H_20		
H/X	H/U	U den.	UO_2F_2 den.	H_2O den.
1.7E+00 % Enriched				
5.000E+00	8.609E-02	4.9325E+00	6.3829E+00	1.6071E-02
1.000E+01	1.722E-01	4.9049E+00	6.3473E+00	3.1963E-02
2.000E+01	3.443E-01	4.8498E+00	6.2760E+00	6.3209E-02
5.000E+01	8.609E-01	4.6845E+00	6.0621E+00	1.5264E-01
1.000E+02	1.722E+00	4.4090E+00	5.7056E+00	2.8732E-01
2.000E+02	3.443E+00	3.8581E+00	4.9926E+00	5.0283E-01
3.000E+02	5.165E+00	2.8746E+00	3.7199E+00	5.6197E-01
5.000E+02	8.609E+00	2.0901E+00	2.7048E+00	6.8103E-01
7.000E+02	1.205E+01	1.6421E+00	2.1249E+00	7.4905E-01
1.000E+03	1.722E+01	1.2425E+00	1.6079E+00	8.0970E-01
1.500E+03	2.583E+01	8.8401E-01	1.1440E+00	8.6411E-01
2.000E+03	3.443E+01	6.8606E-01	8.8781E-01	8.9416E-01
1.4E+00 % Enriched				
5.000E+00	7.090E-02	4.9373E+00	6.3891E+00	1.3248E-02
1.000E+01	1.418E-01	4.9146E+00	6.3598E+00	2.6374E-02
2.000E+01	2.836E-01	4.8693E+00	6.3011E+00	5.2261E-02
5.000E+01	7.090E-01	4.7331E+00	6.1249E+00	1.2700E-01
1.000E+02	1.418E+00	4.5063E+00	5.8314E+00	2.4183E-01
2.000E+02	2.836E+00	4.0525E+00	5.2442E+00	4.3495E-01
3.000E+02	4.254E+00	3.1917E+00	4.1303E+00	5.1384E-01
5.000E+02	7.090E+00	2.3763E+00	3.0751E+00	6.3761E-01
7.000E+02	9.925E+00	1.8927E+00	2.4493E+00	7.1101E-01
1.000E+03	1.418E+01	1.4501E+00	1.8765E+00	7.7819E-01
1.500E+03	2.127E+01	1.0434E+00	1.3503E+00	8.3992E-01
2.000E+03	2.836E+01	8.1489E-01	1.0545E+00	8.7461E-01

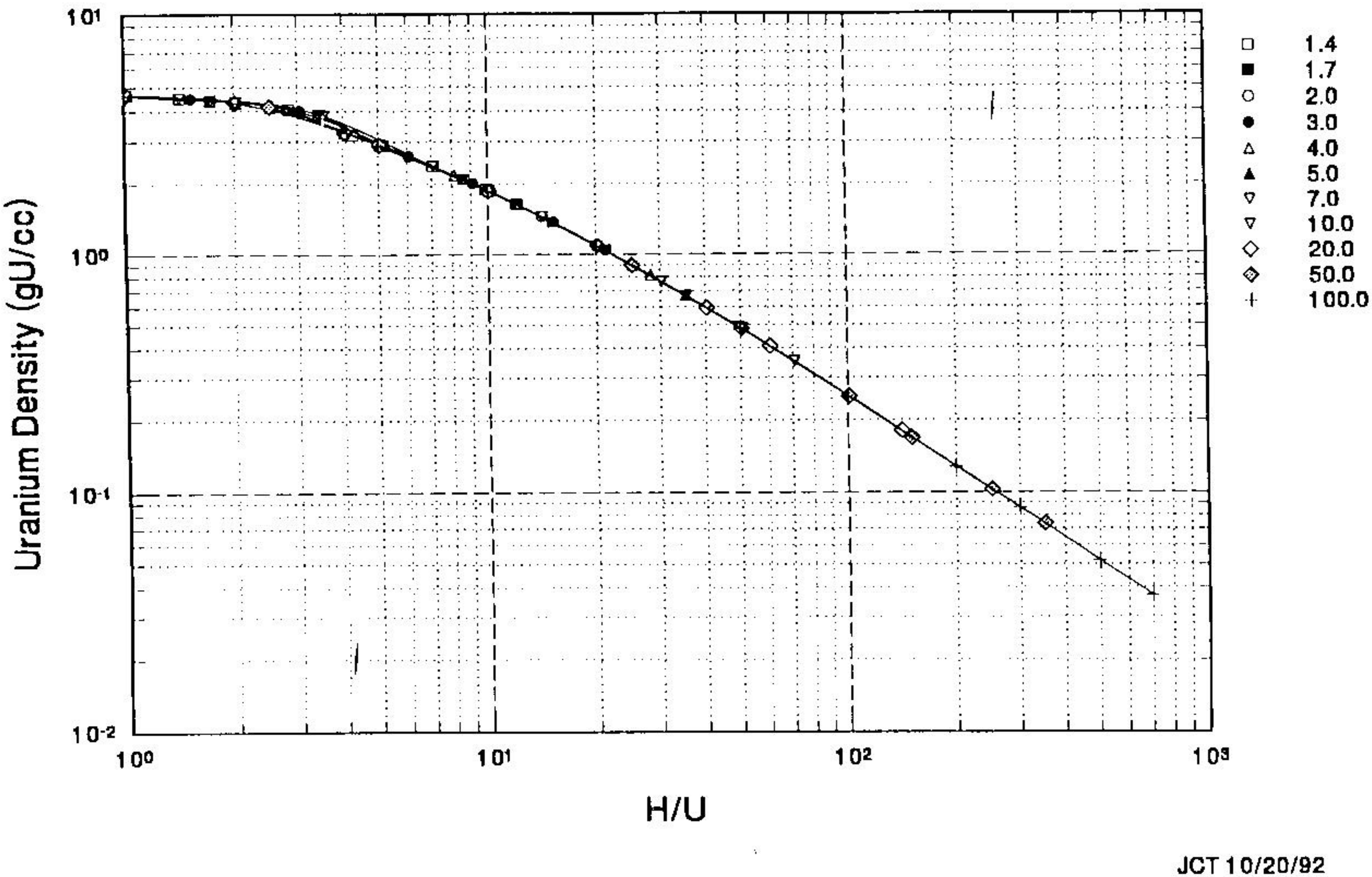

Fig. A.1. Uranium density vs H/U for UO_2F_2–H_2O.

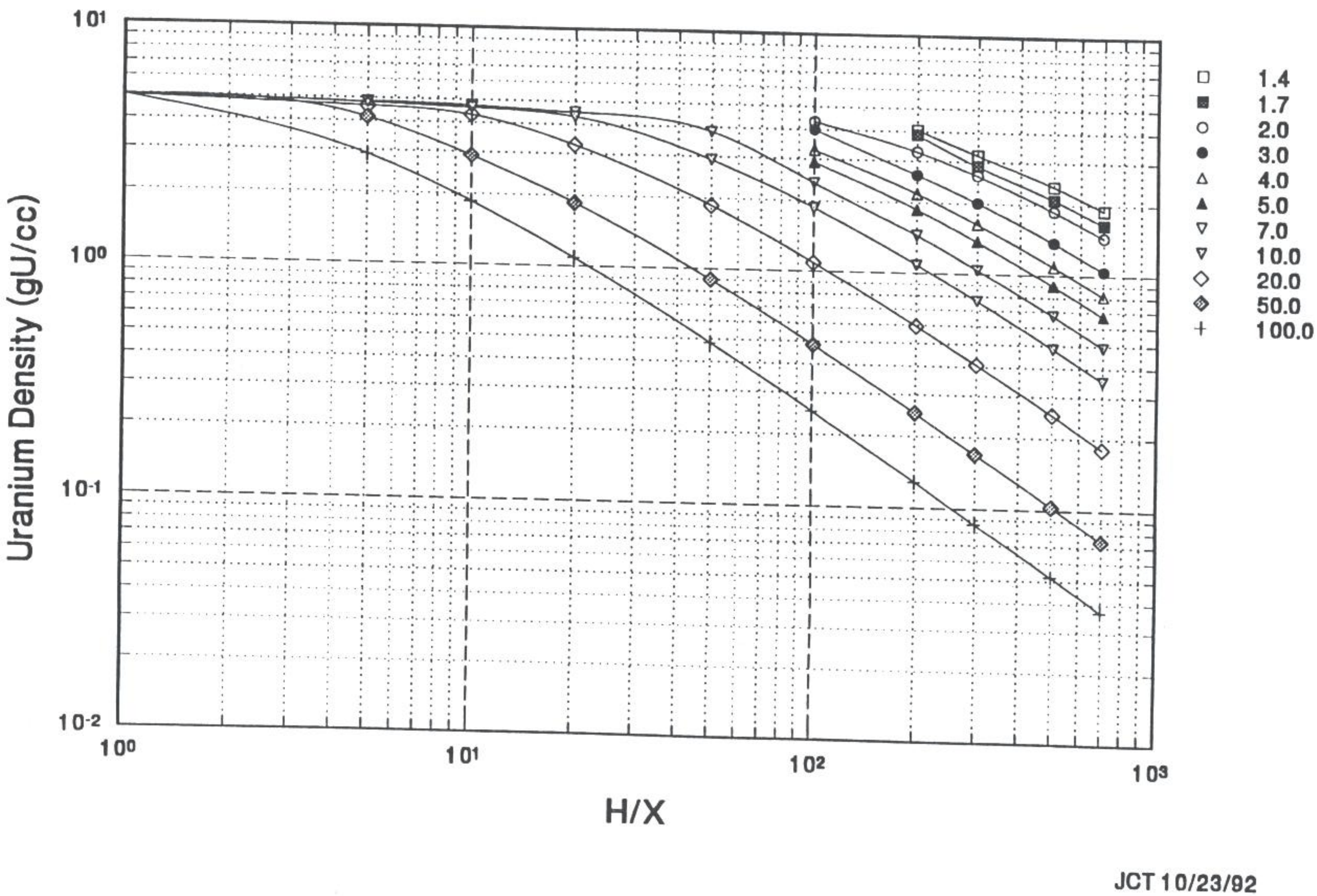

Fig. A.2. Uranium density vs H/X for UO_2F_2–H_2O.

APPENDIX B

Table B.1. k_4 for UO_2F_2–H_2O systems
SCALE 27-group library

	% Enrichment				
H/X	100.0	50.0	20.0	10.0	7.0
0.0	2.08054	1.80595	1.37262	1.04620	0.894432
5.0	1.80732	1.64057	1.40257	1.15864	1.01704
10.0	1.80661	1.64316	1.43940	1.22586	1.09507
20.0	1.83945	1.68521	1.50977	1.32515	1.20771
50.0	1.88762	1.76097	1.62091	1.47394	1.37764
100.0	1.88964	1.79156	1.67708	1.55770	1.47809
200.0	1.83467	1.76613	1.67553	1.58147	1.51812
300.0	1.76675	1.71408	1.63703	1.55673	1.50241
500.0	1.63535	1.59985	1.53996	1.47603	1.43267
700.0	1.51871	1.49250	1.44376	1.39031	1.35396
1000.0	1.37031	1.35205	1.31459	1.27190	1.24267
1500.0	1.17739	1.16580	1.13979	1.10854	1.08686
2000.0	1.03163	1.02356	1.00442	0.980406	0.963521

	% Enrichment					
H/X	5.0	4.0	3.0	2.0	1.7	1.4
5.0	0.881333	0.793512	0.687041	0.555748	0.510421	0.462036
10.0	0.963163	0.873875	0.760571	0.613581	0.560951	0.503967
20.0	1.08401	0.996199	0.879551	0.716552	0.654776	0.585342
50.0	1.27189	1.19358	1.08394	0.916725	0.847631	0.765433
100.0	1.38902	1.32164	1.22466	1.07018	1.00338	0.920974
200.0	1.44607	1.39075	1.30964	1.17623	1.11675	1.04172
300.0	1.44016	1.39206	1.32102	1.20227	1.14849	1.07998
500.0	1.38267	1.34382	1.28586	1.18737	1.14215	1.08382
700.0	1.31182	1.27903	1.22992	1.14571	1.10666	1.05588
1000.0	1.20866	1.18222	1.14238	1.07355	1.04134	0.999196
1500.0	1.06147	1.04178	1.01201	0.960152	0.935683	0.903401
2000.0	0.943632	0.928235	0.904946	0.864176	0.844828	0.819175

Table B.2. Critical parameters for UO_2F_2–H_2O
SCALE 27-group library

Enrichment	H/X	Crit. Rad. cm	Crit. Vol. l	Crit. Mass kg U	Mass H_2O kg
100.0	0	15.020	14.1929	70.3970	.---
	5	11.623	6.5769	19.0143	3.6432
	10	11.400	6.2057	11.5386	4.4217
	20	11.154	5.8135	6.3075	4.8342
	50	11.062	5.6702	2.7349	5.2403
	100	11.367	6.1528	1.5410	5.9055
	200	12.252	7.7039	0.9837	7.5396
	300	13.215	9.6674	0.8284	9.5237
	500	15.298	14.9960	0.7751	14.8513
	700	17.830	23.7422	0.8785	23.5667
50.0	0	21.449	41.3348	205.021	.---
	5	14.398	12.5014	51.9423	4.9767
	10	14.008	11.5147	33.3820	6.3968
	20	13.087	9.3888	17.4874	6.7020
	50	12.266	7.7294	6.9462	6.6554
	100	12.237	7.6751	3.7033	7.0964
	200	12.893	8.9778	2.2489	8.6191
	300	13.788	10.9802	1.8574	10.6778
	500	15.831	16.6184	1.7044	16.3299
	700	18.256	25.4862	1.8755	25.1564
20.0	0	39.228	252.866	1254.2	.---
	5	25.904	72.8055	337.5793	12.9375
	10	20.596	36.5979	157.8640	12.1000
	20	17.421	22.1482	72.3880	11.0969
	50	14.752	13.4478	25.0748	9.6099
	100	13.880	11.2020	12.1733	9.3308
	200	14.128	11.8125	6.9979	10.7277
	300	14.901	13.8588	5.6433	12.9767
	500	16.943	20.3739	5.1045	19.5626
	700	19.462	30.8791	5.5870	29.9765
10.0	0	138.378	11099.15	55051.8	.---
	5	53.907	656.17	3148.4	60.3283
	10	38.217	233.815	1084.0	41.5466
	20	25.822	72.1187	311.022	23.8396
	50	18.735	27.5459	80.0379	15.3373
	100	16.417	18.534	34.5714	13.2494
	200	15.908	16.8624	18.3284	14.0487
	300	16.461	18.6841	14.3292	16.4749
	500	18.474	26.4085	12.7471	24.4265
	700	21.145	39.6043	13.9475	37.4178

Table B.2. (continued)

Enrichment	H/X	Crit. Rad. cm	Crit. Vol. l	Crit. Mass kg U	Mass H_2O kg
7.0	0	.---	.---	.---	.---
	5	205.297	36244.02	175662.8	2356.22
	10	69.819	1425.63	6747.97	181.026
	20	37.903	228.10	1027.99	55.1556
	50	20.772	37.544	143.6699	19.2714
	100	18.661	27.2216	64.6775	17.3513
	200	17.459	22.2919	32.3252	17.3440
	300	17.792	23.5935	24.6191	19.8138
	500	19.727	32.1553	21.4971	28.8352
	700	22.433	47.2867	23.2565	43.6740
5.0	100	22.078	45.078	130.970	25.113
	200	19.621	31.641	59.004	22.627
	300	19.705	32.049	44.003	25.312
	500	21.423	41.184	37.020	35.492
	700	24.384	60.730	40.578	54.464
4.0	100	25.478	69.276	226.588	34.754
	200	21.696	42.779	93.126	28.567
	300	21.384	40.960	66.818	30.745
	500	23.020	51.098	55.523	42.580
	700	26.233	75.619	61.601	66.137
3.0	100	30.860	123.105	490.906	56.463
	200	25.709	71.178	186.088	42.806
	300	24.660	62.816	126.210	43.549
	500	26.125	74.689	102.578	58.989
	700	29.608	108.722	113.419	91.318
2.0	100	68.944	1372.708	5918.842	453.790
	200	37.622	223.056	729.772	111.901
	300	33.551	158.199	413.643	95.141
	500	34.222	167.882	313.184	120.057
	700	39.136	251.083	364.070	195.390
1.7	200	44.494	368.972	1423.531	185.530
	300	40.725	282.925	813.296	158.995
	500	40.492	278.097	581.251	189.392
	700	47.253	441.954	725.733	331.046
1.4	200	83.483	2437.153	9876.563	1060.040
	300	59.670	889.932	2840.396	457.283
	500	55.461	714.581	1698.059	455.624
	700	68.124	1324.308	2506.518	941.569

APPENDIX C

Table C.1. k_4 for various pure compounds @ H/U = 0
SCALE 27-group library

Enrichment	U metal	UO_2	U_3O_8	UO_2F_2	UF_6	$UO_2(NO_3)_2$
1	0.502272	0.430478	0.419889	0.366772	0.312843	0.322399
2	0.643246	0.559364	0.550488	0.495281	0.440475	0.447926
3	0.766336	0.668193	0.659211	0.600001	0.543838	0.549150
5	0.970126	0.846011	0.834575	0.765520	0.705737	0.708462
7	1.13160	0.988147	0.973347	0.894357	0.830416	0.831498
10	1.31911	1.15789	1.13826	1.04615	0.975925	0.974639
12	1.41814	1.25060	1.22825	1.12884	1.05469	1.05157
13	1.46172	1.29221	1.26867	1.16603	1.09003	1.08597
16	1.57383	1.40187	1.37538	1.26454	1.18349	1.17634
20	1.69078	1.52064	1.49147	1.37263	1.28596	1.27456
30	1.88701	1.73076	1.69887	1.56978	1.47342	1.45271
50	2.09617	1.96765	1.93674	1.80597	1.70073	1.66883
70	2.21167	2.10053	2.07201	1.94665	1.83839	1.80160
100	2.31977	2.22252	2.19675	2.08060	1.97151	1.93237

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