Nuclear Criticality Safety Engineer Training
Module 5 [1]

Criticality Limits

LESSON OBJECTIVE

To investigate subcritical limit curves, understand the physics of the curves and consider how various criticality control parameters are illustrated in the curves.

SUBCRITICAL LIMIT CURVES

The following subcritical limit curves from TID-7016 (Nuclear Safety Guide, TID-7016 Rev 2, J. T. Thomas, editor, NUREG/CR-0095 and ORNL/NUREG/CSD-6, 1978) are included at the end of this module (original Figure numbers retained).

1) Figure 2.1. Subcritical mass limits for individual spheres of homogeneous water-reflected and -moderated ^{235}U
2) Figure 2.2. Subcritical volume limits for individual spheres of homogeneous water-reflected and -moderated ^{235}U
3) Figure 2.3. Subcritical diameter limits for individual cylinders of homogeneous water-reflected and -moderated ^{235}U
4) Figure 2.4. Subcritical thickness limits for individual slabs of homogeneous water-reflected and -moderated ^{235}U

Similar curves are included in TID-7016 Rev. 2 for ^{233}U and ^{239}Pu and every criticality specialist should be familiar with them and understand them. The curves in this module are for single units of the material, either as ^{235}U metal homogeneously mixed with water or for $^{235}UO_2F_2$ solutions. These curves are calculated, not measured; and although they were calculated with the codes and cross section sets in use several decades ago, they are still valid and widely used to develop criticality control parameter limits.

Figure 2.1 of TID-7016 Rev. 2 is a plot of the spherical ^{235}U subcritical mass limit in kg ^{235}U versus the uranium concentration in kg(^{235}U)/L. There are also insert scales with the hydrogen-to-fissile (H:U) atomic ratio for the metal-water mixture and the UO_2F_2 solution. The curves on the figure represent subcritical limits; i.e., mixtures below the line would be subcritical and those above the line supercritical at any given concentration. There are two sets of curves on the figure, one set for metal-water mixtures and one set for UO_2F_2 solutions. Each set has a curve for a thin

[1] Developed for the U. S. Department of Energy Nuclear Criticality Safety Program by T. G. Williamson, Ph.D., Westinghouse Safety Management Solutions, Inc., in conjunction with the DOE Criticality Safety Support Group.

(25 mm) and a thick (300 mm) water reflector. The curves are terminated on both ends of the figure. At high uranium concentrations, the metal curves end at 18.9 kg(^{235}U)/L, which is the metal density, and the solution curves end at about 1.3 kg(^{235}U)/L, the density of UO_2F_2 at its solubility limit. At the lower end of the concentration scale the four curves come together and appear to be rising to an asymptotic concentration limit.

Figures 2.2 through 2.4 of TID-7016 Rev. 2 display similar curves for volume, cylinder diameter and infinite slab thickness subcritical limits as a function of uranium concentration.

Next consider some values that can be read from these figures and compare them to the corresponding values in ANSI/ANS-8.1 (American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors, ANSI/ANS-8.1-1998) for ^{235}U. Table 1 below summarizes some of the single parameter subcritical limits than can be read from both the Standard and the curves. (**Note**: Others may read the curves differently, and the values from TID-7016 Rev. 2 should be considered only approximate.) It appears to be a reasonable conclusion that these curves and the values in the ANSI/ANS-8.1 Standards are related.

Table 1. Single Parameter ^{235}U Subcritical Limits

Parameter	Source	Subcritical Limit
Metal unit mass	• ANS-8.1, Table 3 • TID-7016, Fig. 2.1, metal-water curve at H:U = 0	20.1 kg 18 kg
Solution concentration	• ANS-8.1, Table 1 • TID-7016, Fig. 2.1, low concentration asymptote	11.6 g/L 13 g/L
Atomic ratio, H:F	• ANS-8.1, Table 1 • TID-7016, Fig. 2.1, low concentration asymptote	2250 2000
Aqueous mixtures	• ANS-8.1, Section 5.2 • TID-7016, Fig. 2.1, metal-water curve minimum	0.70 kg 0.65 kg
Volume of solution (UO_2F_2)	• ANS-8.1, Table 1 • TID-7016, Fig. 2.2, minimum of (UO_2F_2) curve	5.5 L 4.8 L
Diameter of cylinder of solution (UO_2F_2)	• ANS-8.1, Table 1 • TID-7016, Fig. 2.3, minimum of (UO_2F_2) curve	13.7 cm 13 cm
Diameter of metal cylinder	• ANS-8.1, Table 3 • TID-7016, Fig. 2.3, metal-water curve at H:U = 0	7.3 cm 7.0 cm
Thickness of slab of solution (UO_2F_2)	• ANS-8.1, Table 1 • TID-7016, Fig. 2.4, minimum of (UO_2F_2) curve	4.4 cm 4.2 cm
Thickness of metal slab	• ANS-8.1, Table 3 • TID-7016, Fig. 2.4, metal-water curve at H:U = 0	1.3 cm 1.25 cm

Consider the shape of the subcritical mass curves in Fig. 2.1. The lower curve is for a 300-mm thick water reflector, which is normally considered an infinitely thick reflector. That is, increasing the reflector thickness will not change the reactivity of the system; or put another way, if a neutron were to get 30 cm from the fissile material, the chances of it returning to the fissile sphere are negligibly small. Remember that the neutron mean free path for scattering in water is less than a centimeter.

Look at the high-concentration end of the fully-reflected metal system. There is negligible moderation of the neutrons in the metal so the neutrons either undergo fast capture in the metal, which may be a fission event, or escape to the water reflector. In the reflector the neutrons can be moderated to lower energies. In this process they can either be absorbed in the water, scattered back into the fissile solution or scattered out of the reflector. One likely possibility is that some neutrons will be thermalized in the reflector and cause thermal fission in the outer few millimeters of the metal. Note that the difference in subcritical mass between a thick reflector, 30 cm, and a thin one, 2.5 cm, is about 10 kg of ^{235}U. This indicates that with a thick reflector a fair number of neutrons, which otherwise might be lost, will be returned to participate in the chain reactions.

As hydrogen is added to the metal system nothing much happens to the shape of the curve until H:U increases to about 3. If anything, the subcritical mass rises slightly because hydrogen increases the scattering and the leakage and a few neutrons are scattered from the fast fission region to the resonance region where neutron capture resonances may be more important than fission resonances. As the hydrogen concentration increases further, the subcritical mass limit drops sharply. The neutrons now become adequately moderated in the mixture and the average fission energy shifts into the thermal energy region. The mass curve comes to a minimum then increases as the additional water is included in the solution. In this region the additional water does not add to the moderation but absorbs neutrons and acts like a neutron poison. The minimum of the curve is a region of optimum moderation and systems are often spoken of as being over-moderated on one side of the minimum and under-moderated on the other. At the lower end of the uranium concentration scale, the subcritical mass curve rises sharply and the uranium becomes so dilute that criticality is impossible.

This curve illustrates some basic criticality control concepts:

- moderation - as the hydrogen concentration changes the subcritical limit changes
- concentration - there is a low concentration limit
- reflection - different curves for 25 and 300 mm water reflector
- geometry - spherical geometry gives the lowest critical mass
- absorption - at high hydrogen concentrations the hydrogen absorption causes the subcritical mass limit to increase
- mass - the curve provides minimum subcritical mass limit values.

This curve does not illustrate the effects of:

- enrichment - this curve is for ^{235}U; ^{238}U in the mixture would move the curves up
- spacing - this curve is for a single unit
- temperature - there is no temperature dependence here.

These curves are called subcritical limit curves. In the area of the graph above the curves the system is supercritical and below the curves the system is subcritical. Does this mean that if you were to calculate the conditions representing a point on the line you would calculate $k_{eff} = 1.00$? Probably not. You would probably get a value a few per cent below 1.0. Two reasons are suggested:

1) We do not know what the originators of this curve defined as subcritical. That is, we do not know what bias or margin was applied to these calculations. It is likely that a 2-3% margin was used.

2) If we did the calculation with today's computational tools we would be using a different computing platform and different cross sections and should not expect to get the same answer.

Other references such as "Critical Dimensions of Systems Containing ^{235}U, ^{239}Pu and ^{233}U, 1986 Revision," (LA-10860-MS, Los Alamos National Laboratory) contain critical configuration data that can be compared to the subcritical limits of TID-7016 Rev. 2.

EXAMPLE

Calculate the conditions of a uranium-water system near the minimum of the 300-mm reflected curve where the ^{235}U concentration is 0.05 kg/L and the ^{235}U mass is 0.6 kg. To calculate the solution characteristics use the following density values:

uranium metal	18.9 g/cm^3
water	0.9982 g/cm^3 at 20^o C.

To find the density of water in the solution use the sum of the fractional volumes.

$$1 = \sum \frac{\rho^i}{\rho_0^i} = \frac{0.05}{18.9} + \frac{water}{0.9982}$$

where ρ^i is the density of component i in the solution and ρ_0^i is the natural density of that component. With this calculation we get

water	0.9956	g/cm^3 in solution
^{235}U	0.050	g/cm^3 in solution
solution (sum)	1.0456	g/cm^3.

The volume of the sphere is

600 g ^{235}U/(0.050 g/cm^3) = 12000 cm^3

and the sphere radius is 14.20 cm.

The atom densities and H:U ratio are

^{235}U	1.2811×10^{-4} at/b-cm
H	6.6558×10^{-2} at/b-cm
O	3.3279×10^{-2} at/b-cm
H:U	520

These values can be input to a modern computational tool and it is suggested that each criticality engineer make this calculation. First guess what k_{eff} the code will compute based on the curves in TID-7016 Rev. 2 and test your intuition about the subcritical nature of the curves.

A typical result should be similar to the one from a short KENO-Va calculation using the 238-group ENDF/B-V cross section set, which yields k_{eff} of 0.94.

SUMMARY

In this module the single-parameter subcritical mass limit was introduced as displayed on the curves in TID-7016 Rev. 2, and as given in the Standard ANSI/ANS-8.1-1998. Both tools can be useful in setting criticality control parameter limits for simple systems provided the user understands them and does not apply them to systems outside their range of applicability.

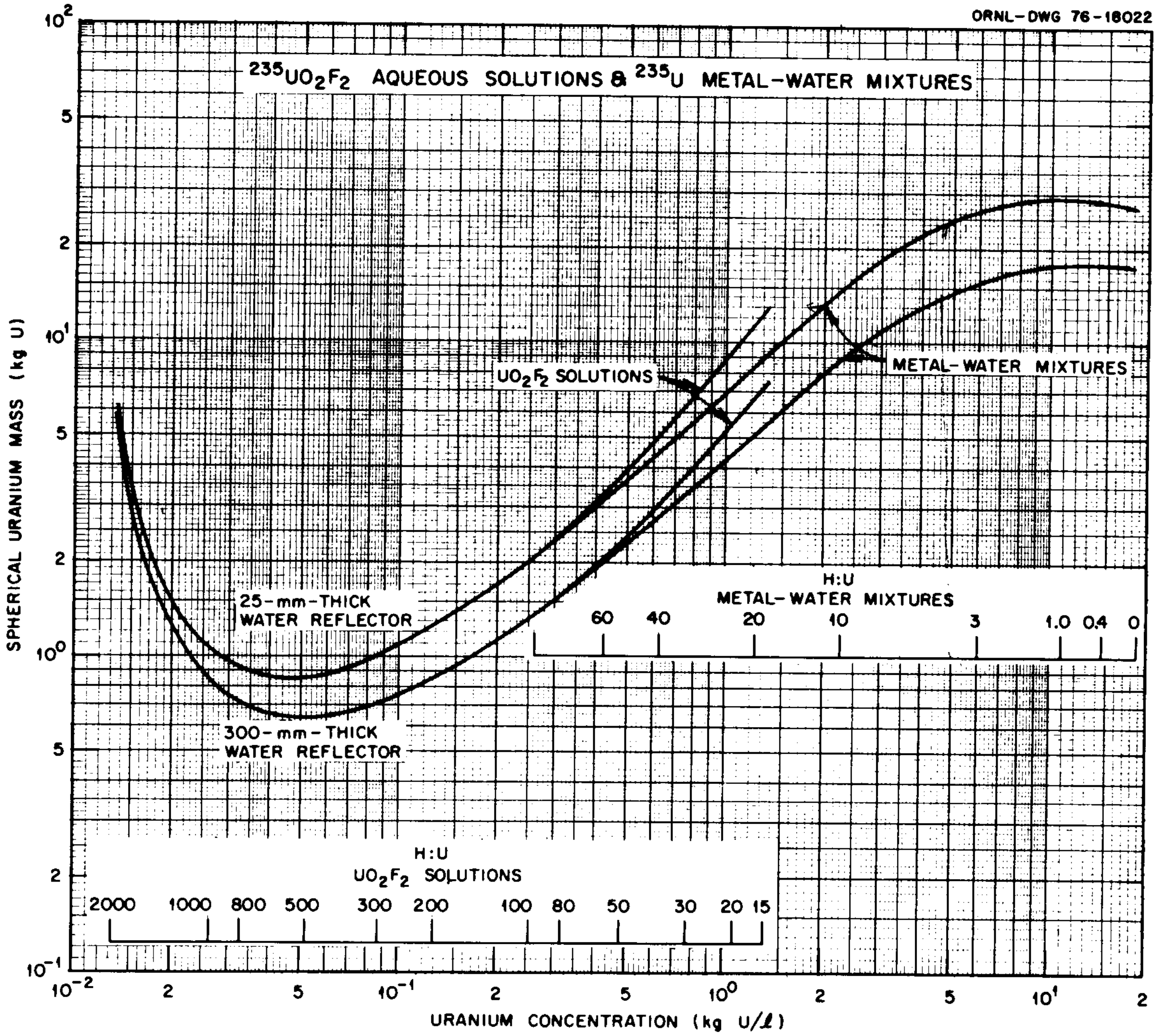

Figure 2.1. Subcritical mass limits for individual spheres of homogeneous water-reflected and -moderated ^{235}U.

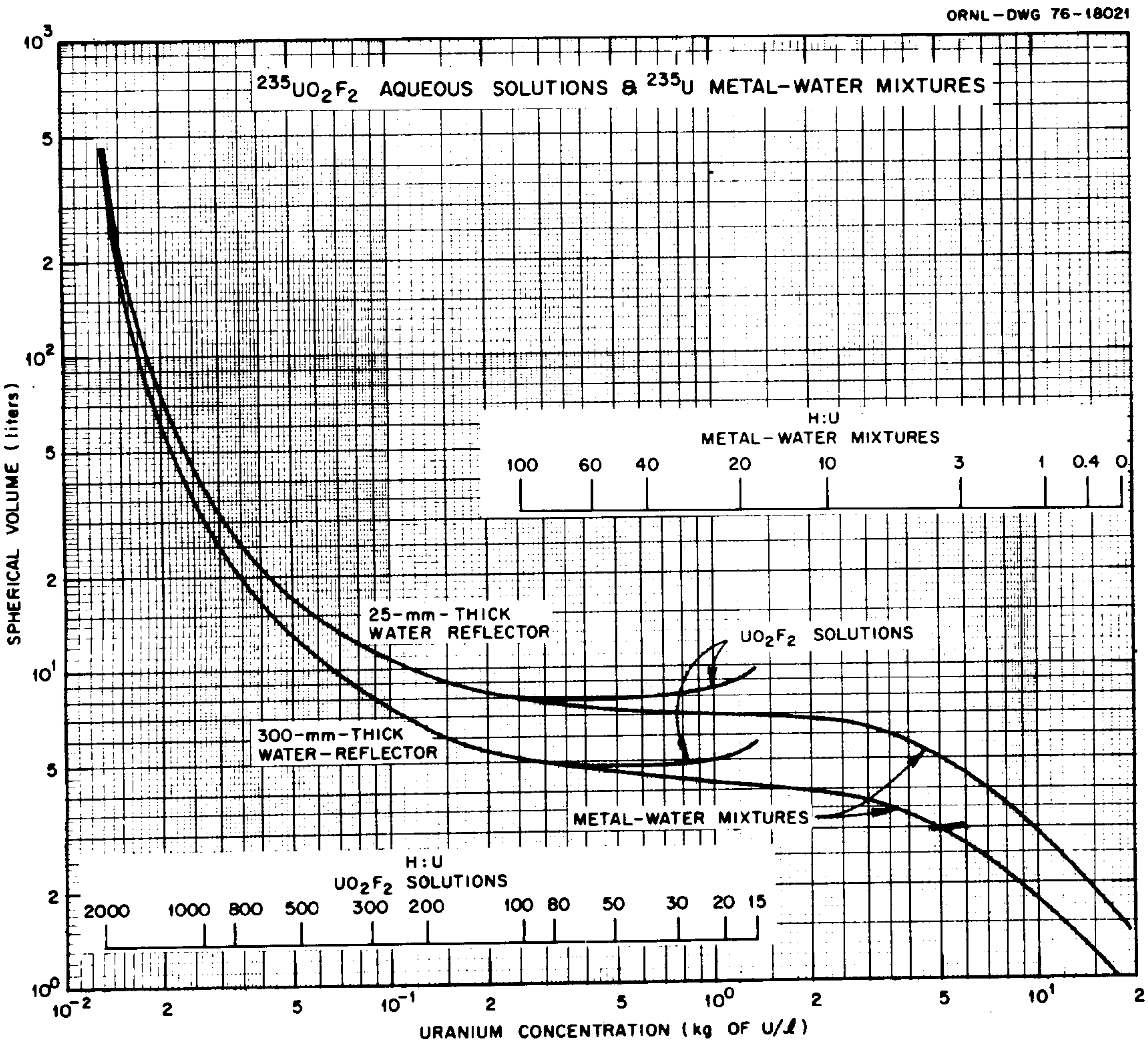

Figure 2.2. Subcritical volume limits for individual spheres of homogeneous water-reflected and -moderated ^{235}U.

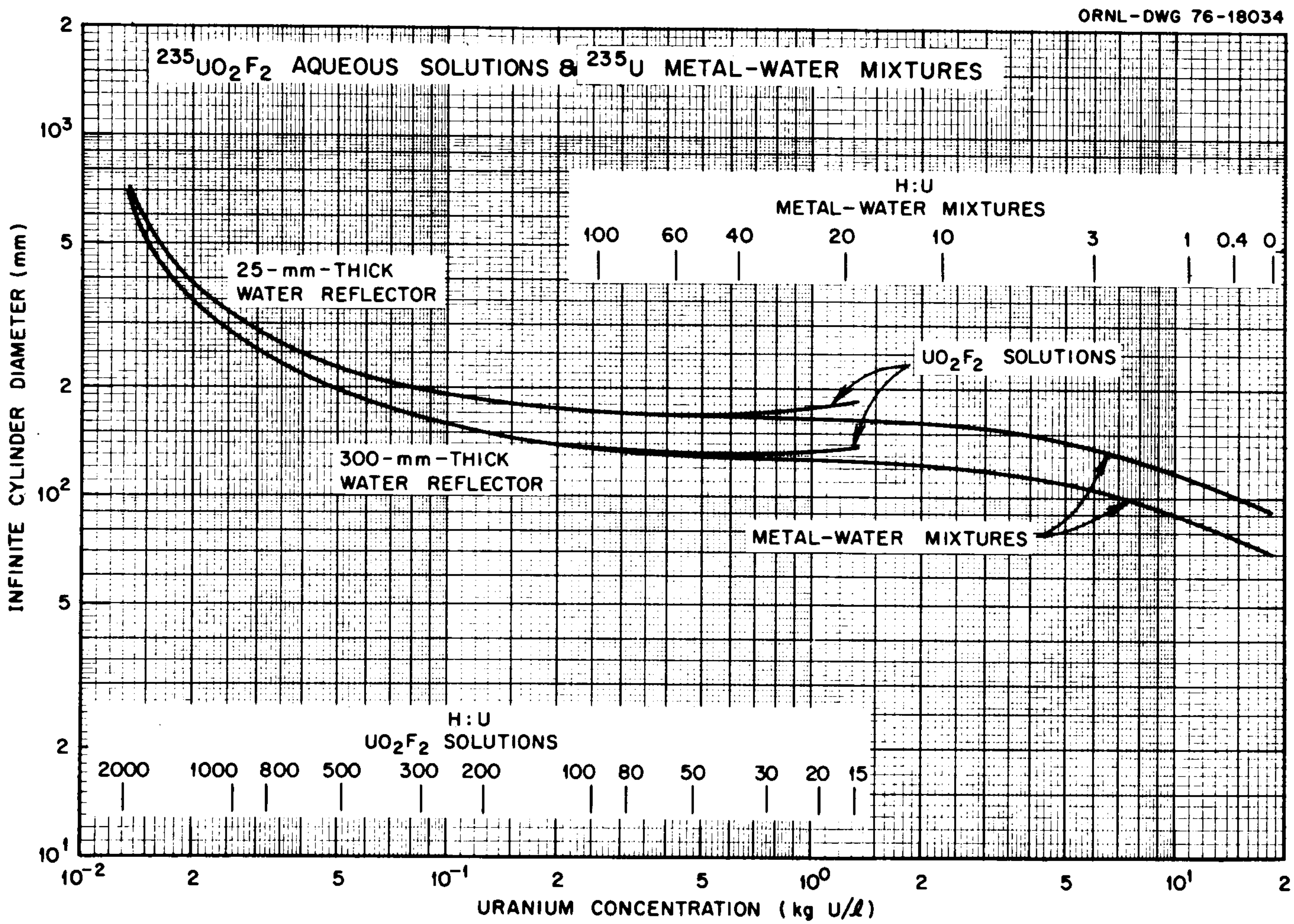

Figure 2.3. **Subcritical diameter limits for individual cylinders of homogeneous water-reflected and -moderated ^{235}U.**

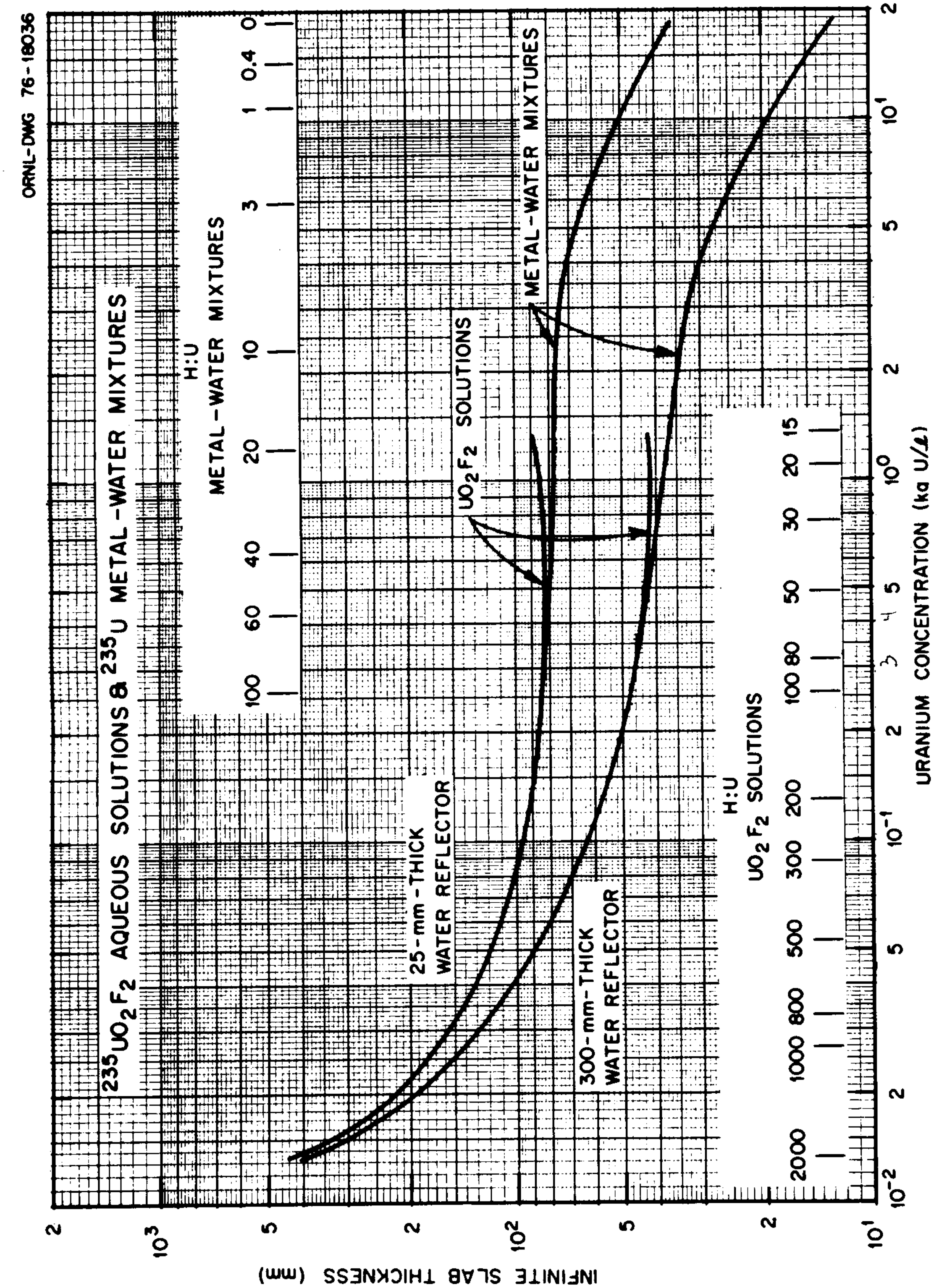

Figure 2.4. **Subcritical thickness limits for individual slabs of homogeneous water-reflected and -moderated ^{235}U.**

PROBLEMS

1. Below is a copy of the subcritical mass limit curves for ^{239}Pu metal and oxide in water mixtures from TID-7016 Rev. 2. Read the following values from these curves and compare the results to the values in ANSI/ANS-8.1.

- A. What is the minium subcritical mass for a ^{239}Pu water mixture?
- B. What is the minium subcritical mass for a dry metal sphere of ^{239}Pu?
- C. What is the minium subcritical mass for a dry sphere of PuO_2?
- D. What is the plutonium concentration in a water mixture at the minimum subcritical mass?
- E. What is the H:Pu atom ratio in a water mixture at the minimum subcritical mass?
- F. What is the approximate subcritical concentration limit for Pu in water?
- G. What is the H:Pu atom ratio at the minimum subcritical concentration?
- H. What is the difference in subcritical mass for a fully reflected metal sphere and one with a thin reflector?

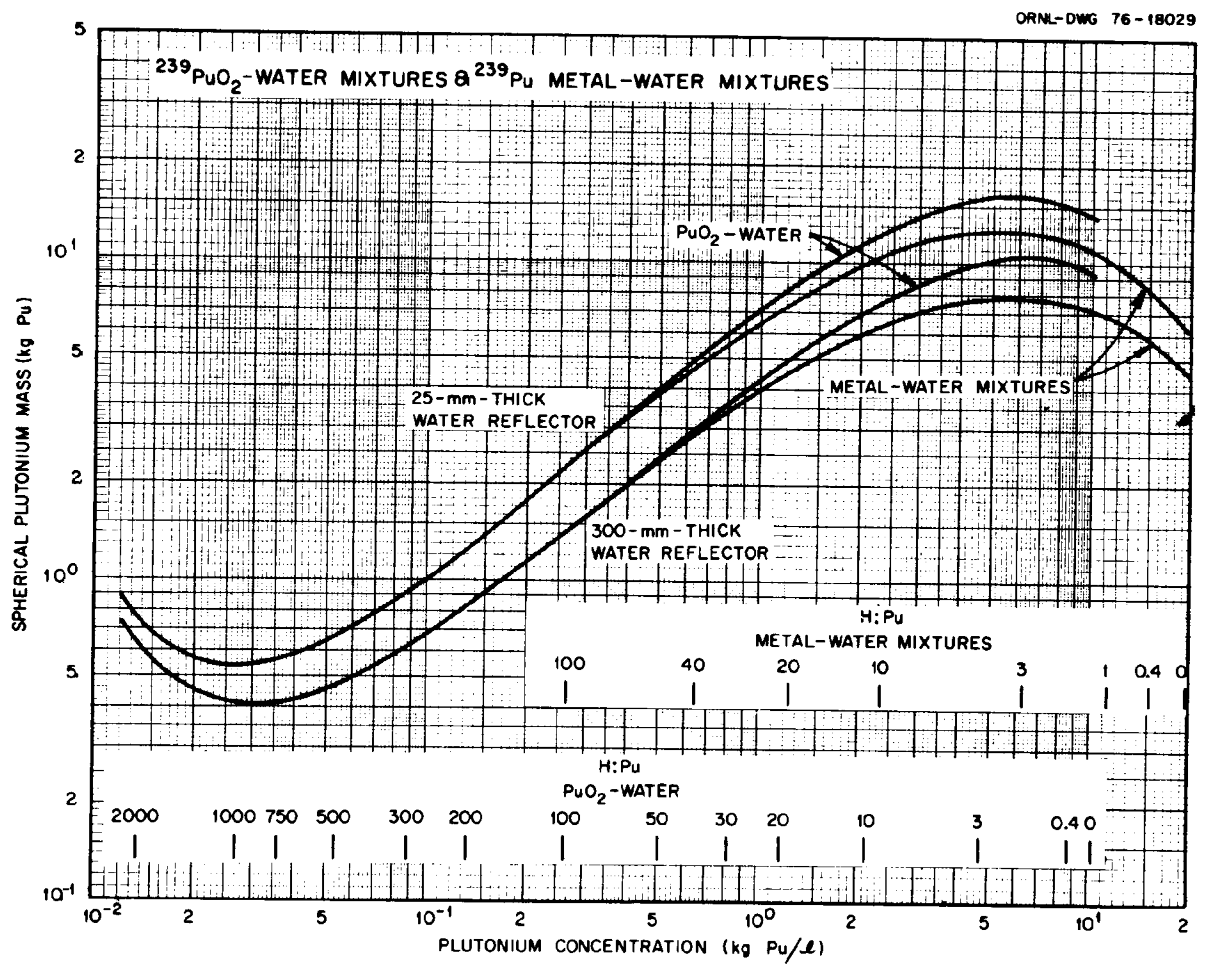

Figure 2.9 Subcritical mass limits for individual spheres of homogeneous water-reflected and -moderated ^{239}Pu.

2. On the plutonium subcritical mass limit curve (see Problem 1) the minimum subcritical mass for a metal-water mixture is 400 g ^{239}Pu at a concentration 30 g(^{239}Pu)/L for a fully water-reflected sphere. Calculate the atom densities of ^{239}Pu, hydrogen and oxygen in this mixture. Calculate the volume and radius of the sphere. If you have access to a criticality code, calculate the multiplication factor of this sphere.

3. On the plutonium subcritical mass limit curve (see Problem 1) the minimum subcritical mass for a metal-water mixture is 400 g ^{239}Pu at a concentration 30 g(^{239}Pu)/L for a fully water-reflected sphere.

A) Use the following thermal neutron parameters to calculate $\nu\Sigma_f / \Sigma_a$ for the mixture. The atom densities calculated in Problem 2 are ^{239}Pu: 7.5575E-05; H: 6.6633E-02; and O: 3.3317E-02 (all atoms/barn-cm). Keep the plutonium concentration at 30 g(^{239}Pu)/L and dilute the ^{239}Pu by adding ^{240}Pu to the mixture. Calculate $\nu\Sigma_f / \Sigma_a$ as a function of the ^{239}Pu fraction.

	^{239}Pu	^{240}Pu	H	O
ν	2.877			
σ_a	1017.3	289.5	0.333	0.00019
σ_f	748.1			
σ_s	7.6	1.64	20.491	3.761

B) For a mixture with 95% ^{239}Pu and 5% ^{240}Pu the atom densities are: ^{239}Pu: 7.1796E-05; ^{240}Pu: 3.7630E-06; H: 6.6633E-02, and O: 3.3317E-02 (all atoms/barn-cm). Using the thermal parameters above, calculate the fraction of the interactions which are absorptions for each component of the mixture and the fraction which are scattering interactions.

4. A homogeneous mixture of water and plutonium at optimum moderation in a spherical balloon is exactly critical, i.e, k_{eff} = 1.00000. This sphere is suspended in the center of a very large empty room. What do you expect to happen to the neutron multiplication of the solution in each of the following cases. Each case in independent of the other situations.

A) A person walks up to the sphere to look at it closely.
B) A steel robot crawls near the sphere to examine it.
C) The sphere is squashed to a pancake shape.
D) The sphere is wrapped in cadmium.
E) A spray of borated water is directed over the sphere. The spray does not penetrate the balloon surface.
F) A neutron source, small in volume but emitting lots of neutrons, moves near the sphere.
G) A similar critical sphere moves to within one foot of the sphere.
H) The sphere develops a leak and the solution splashes on the floor.
I) The system is left alone and ignored for several days.
J) Some of the water evaporates away from the solution.

PROBLEM SOLUTIONS

1.

	TID-7016 Rev. 2	ANSI/ANS-8.1
A	0.4 kg	0.45 kg
B	4.5 kg	5.0 kg
C	9 kg	10.2 kg
D	0.03 kg/L = 30 g/L	
E	800	
F	0.01 kg/l = 10 g/L	7.3 g/L (solution only)
G	2500	3630 (solution only)
H	6 kg - 4.5 kg = 1.5 kg	

2. To calculate the solution characteristics use these values: plutonium metal density = 19.6 g/cm^3; water density = 0.9982 g/cm^3 at 20 ^{o}C. Following the example on Page 4, use the sum of fractional densities to calculate the density of water in the solution.

$$1 = \sum \frac{\rho^i}{\rho_0^i} = \frac{0.030}{19.6} + \frac{\text{water}}{0.9982}$$

This gives

water	0.9967 g/cm^3 in solution
^{239}Pu	0.030 g/cm^3 in solution
solution	1.0267 g/cm^3.

The volume of the sphere is

400 g ^{239}Pu/(0.030 g/cm$^{3)}$ = 13333 cm^3

and the sphere radius is 14.71 cm.

Using standard values for the atomic weights, the atom densities are calculated to be (at/b-cm)

^{239}Pu	7.5575 x 10^{-5}
H	6.6633 x 10^{-2}
O	3.3317 x 10^{-2}
H:Pu	882

Here is a sample input file for SCALE 4.3 using KENO-Va with the 27-group ENDF/B-IV cross section set.

```
=csas25
plutonium aqueous sphere
27groupndf4  infhommedium
pu-239    1 0.0 7.5575e-05  298 end
h         1 0.0 6.6633E-02  298 end
o         1 0.0 3.3317E-02  298  end
h         2 0.0 6.6735E-02  298  end
```

```
o         2 0.0 3.3368E-02  298  end
end comp
plutonium aqueous sphere
read parm gen=500 npg=500 nsk=10 end parm
read geom
sphere  1  1  14.71
sphere  2  1  44.71
end geom
end data
end
```

The computed k-effective is 0.96050 +/ - 0.00170.

Here is a sample input file for MCNP4A using ENDF/B-V cross sections.

```
reflected pu sphere 30 g pu239/l
C 400 g Pu239
C cell cards
1   1  1.0003e-01    -1   imp:n=1 $solution sphere
2   2  1.0010e-01     1  -2  imp:n=1 $water reflector
3   0           2        imp:n=0    $outside world
C surface cards
1 so 14.71          $ sphere radius
2 so 44.71          $ water reflector
C control cards
kcode  500 1.0 10 510
ksrc    0 0 0
m1      94239.50c 7.5575e-05
        1001.50c 6.6633e-02
        8016.50c 3.3317e-02
mt1     lwtr
m2     1001.50c 6.6735e-02
       8016.50c 3.3368e-02
mt2    lwtr
```

The computed keff = 0.95588 with an estimated standard deviation of 0.00158. We might conclude that the generators of the curve in TID-7016 Rev. 2 used a combination of bias and margin of about 0.04 in k_{eff}.

3. (A) This problem is similar to one worked in Module 3. Note that the plutonium concentration is a mass density, so the atom densities in the problem must be recalculated as the ratio of ^{239}Pu to ^{240}Pu changes. The macroscopic cross sections are the products of the atom densities and the microscopic cross sections. The following table shows the results for ^{239}Pu fractions from 1.0 to 0.1.

f(^{239}Pu)	N(^{239}Pu)	N(^{240}Pu)	$\nu\Sigma_f$	Σ_a	$\nu\Sigma_f / \Sigma_a$
1	7.5575e-05	0.0000e+00	1.6266e-01	9.9077e-02	1.642
0.95	7.1796e-05	3.7630e-06	1.5453e-01	9.6323e-02	1.604
0.9	6.8017e-05	7.5259e-06	1.4639e-01	9.3568e-02	1.565
0.85	6.4239e-05	1.1289e-05	1.3826e-01	9.0813e-02	1.522
0.8	6.0460e-05	1.5052e-05	1.3013e-01	8.8058e-02	1.478
0.75	5.6681e-05	1.8815e-05	1.2199e-01	8.5304e-02	1.430
0.7	5.2902e-05	2.2578e-05	1.1386e-01	8.2549e-02	1.379
0.65	4.9124e-05	2.6341e-05	1.0573e-01	7.9794e-02	1.325
0.6	4.5345e-05	3.0104e-05	9.7595e-02	7.7039e-02	1.267
0.55	4.1566e-05	3.3867e-05	8.9462e-02	7.4285e-02	1.204
0.5	3.7787e-05	3.7630e-05	8.1329e-02	7.1530e-02	1.137
0.45	3.4009e-05	4.1393e-05	7.3196e-02	6.8775e-02	1.064
0.4	3.0230e-05	4.5156e-05	6.5063e-02	6.6021e-02	0.986
0.35	2.6451e-05	4.8919e-05	5.6930e-02	6.3266e-02	0.900
0.3	2.2672e-05	5.2682e-05	4.8797e-02	6.0511e-02	0.806
0.25	1.8894e-05	5.6445e-05	4.0665e-02	5.7756e-02	0.704
0.2	1.5115e-05	6.0208e-05	3.2532e-02	5.5002e-02	0.591
0.15	1.1336e-05	6.3970e-05	2.4399e-02	5.2247e-02	0.467
0.1	7.5575e-06	6.7733e-05	1.6266e-02	4.9492e-02	0.329

Remember that these are thermal neutron cross sections only and resonance effects are not included. With this caveat the values indicate that this aqueous mixture could not be critical with less than 40% ^{239}Pu.

(B) Since the macroscopic cross sections are additive, this becomes a simple problem of multiplying atom densities times the microscopic cross sections and forming the ratios. The table below shows the results.

	^{239}Pu	^{240}Pu	H	O	Total	Fraction
Atom density	7.180e-05	3.763e-06	6.663e-02	3.332e-02		
Σ_a	7.304e-02	1.089e-03	2.219e-02	6.330e-06	9.632e-02	6.1%
Σ_s	5.457e-04	6.171e-06	1.365e+00	1.253e-01	1.491e+00	93.9%
Σ_{tot}	7.358e-02	1.096e-03	1.388e+00	1.253e-01	**1.588e+00**	
Fraction abs	75.83%	1.13%	23.04%	0.01%		
Fraction scat	0.04%	0.00%	91.56%	8.40%		
Fraction tot	4.64%	0.07%	87.40%	7.89%		

From the table, 87% of the interactions are with hydrogen, 94% of the interactions are scattering, and 6% are absorption. Of the absorptions, 76% are in ^{239}Pu and 23% in hydrogen.

4.

A) The multiplication would probably increase because of neutrons reflected by the person.
B) The multiplication would probably increase because of neutrons reflected by the robot.
C) The multiplication would probably decrease because of the unfavorable geometry and higher leakage.
D) The multiplication would probably increase because of neutrons reflected by the cadmium. Although cadmium is a good neutron absorber, cadmium on the surface of the sphere would be absorbing leaking neutrons, which in a large room were lost anyway.
E) The multiplication would probably increase because of neutrons reflected by the spray.
F) As long as the source is small enough not to act as a reflector the multiplication probably does not change. The source may add neutrons to the system which may alter the neutron flux level, or power level, but not change the multiplication.
G) If the second sphere reflects neutrons the multiplication would probably increase. If the reflection is not altered then the answer is the same as F.
H) The multiplication would probably decrease as the solution goes to a less favorable geometry on the floor.
I) The multiplication would probably decrease as the fissile material is burned out.
J) The multiplication would probably decrease since the solution was at optimum moderation. However, it is hard to imagine a situation in which water is lost from the solution without changing the geometry and not enough information is given to estimate the geometry change effect on the multiplication.