EPD-CTG-960046

Metal Poisons in Waste Tanks (U)

CONF-961103--22

by
T. G. Williamson
Westinghouse Savannah River Company
Savannah River Site
Aiken, South Carolina 29808

A document prepared for AMERICAN NUCLEAR SOCIETY 1996 INTERNATIONAL CONFERENCE at Washington, DC, USA from 11/10/96 - 11/15/96.

DOE Contract No. **DE-AC09-89SR18035**

This paper was prepared in connection with work done under the above contract number with the U. S. Department of Energy. By acceptance of this paper, the publisher and/or recipient acknowledges the U. S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering this paper, along with the right to reproduce and to authorize others to reproduce all or part of the copyrighted paper.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MASTER

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

This report has been reproduced directly from the best available copy.

Available to DOE and DOE contractors from the Office of Scientific and Technical Information, P.O. Box 62, Oak Ridge, TN 37831; prices available from (615) 576-8401.

Available to the public from the National Technical Information Service, U.S. Department of Commerce, 5285 Port Royal Road, Springfield, VA 22161.

SAFETY ENGINEERING DEPARTMENT
CRITICALITY AND RADIATION TRANSPORT SECTION
CRITICALITY TECHNOLOGY GROUP

EPD-CTG-960046

August 1996

METAL POISONS IN WASTE TANKS

T. G. Williamson
Bldg 730-B, Rm 338
Westinghouse Savannah River Co.
Aiken, SC 29808

A paper for presentation at the
American Nuclear Society
1996 International Conference
Washington, DC
November 10-15, 1996

The information contained in this article was developed during the course of work under Contract N0. DE-AC09-89R18035 with the U.S. Department of Energy. By acceptance of this paper, the publisher and/or recipient acknowledges the U.S. Government's right to retain non-exclusive, royalty-free license in and to any copyright covering this paper along with the right to reproduce, and to authorize other to reproduce all or part of the copyrighted paper.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

METAL POISONS IN WASTE TANKS

INTRODUCTION

Many of the storage tanks with waste from processing fissile materials contain, along with the fissile material, metals which may serve as nuclear criticality poisons. It would be advantageous to the criticality evaluation of these wastes if it can be demonstrated that the poisons remain with the fissile materials and if an always safe poison-to-fissile ratio can be established. The first task, demonstrating that the materials stay together, is the job of the chemist, the second, demonstrating an always safe ratio, is the job of the physicist. The latter task is the object of this paper.

At Savannah River Goslen and others have demonstrated safe ratios for Fe and Mn to U-235, Fe and Mn to Pu-239, and ratios of Cr and Zn to U-235 and Pu-239. In these studies the Hansen Roach 16-group cross sections were used with the Savannah River Site code HRXN in the Joshua J-70 operating system. This code computes the infinite multiplication factor with a B_1 slowing down spectrum. Resonance cross sections are included at 20 values of the potential scattering cross section per atom of absorber and the code uses 3-point Lagrangian interpolation. Goslen et al. computed bias adjusted multiplication factors and defined safe ratios such that the bias adjusted k values were less than 0.95. The results of these studies are summarized in Table 1.

Table 1
Safe Weight Ratios
From Goslen

Mixture	Weight Ratio	Condition
Fe:U-235	77:1	Dry
Fe:Pu-239	160:1	Wet
Mn:U-235	30:1	Dry
Mn:Pu-239	32:1	Dry
Cr:U-235	52:1	Wet
Cr:Pu-239	120:1	Wet
Zn:U-235	165:1	Wet
Zn:Pu-239	350:1	Wet

In this table the weight ratios are the ratio of grams-poison to grams-fissile material and the notation wet or dry refers to the condition of maximum reactivity. Dry means the system is most reactive with no water and wet means the system is most reactive with hydrogen-to-fissile ratio in the range of 100 to 200.

In a companion study, Williamson considered mixtures of Fe, Mn and Cr (cross sections for Zn are not available in several data sets) with U-235 and Pu-239 with other cross section sets. They include ENDF/B-V cross sections processed with the GLASS system on Joshua J-70, ENDF/B-V cross sections in the LAW library processed with AMPX-77, and the default cross sections in MCNP. That study used the safe mixtures from Goslen et al. in both wet and dry conditions and showed that for Fe and Mn the highest multiplication factors were computed with the Hansen Roach cross sections and concluded that the Goslen study is conservative for these mixtures. This was not true for Cr mixtures.

In the Winter 1993 issue of *Criticality Safety Quarterly,* Palmer[1] showed that for certain mixtures of Al, Fe and Zr with U-235 the computed infinite multiplication factor may differ by as much as 20% with different cross sections and processing systems. Parks et al.[2,3] have further studied these mixtures with a variety of cross section sets and processing codes and state "..these metal/uranium mixtures are very sensitive to the metal cross section data in the intermediate-energy range and the processing methods that are used." They conclude with a call for more experimental data.

At WSRC Trumble has analyzed aluminum-uranium mixtures characteristic of research reactor fuel in both the dry and moderated state and shows some scatter in the computed multiplication factors with different cross section for the dry conditions. His scatter is not as great as that reported by Palmer and Parks et al. but he uses a much smaller aluminum to uranium ratio. He also concludes with a call for additional experimental data.

The purpose of this study is to reexamine Goslen's work with cross sections and processing codes used at WSRC today. This study will focus on U-235 mixtures with Fe, Mn, and Cr. Sodium will be included in the list of poisons since sodium is an abundant element in many wastes.

DISCUSSION

Computations were done on the RSK-6000 workstation cluster with SCALE 4.2, SCALE 4.3 and MCNP 4a. The 27-group ENDF/B-IV cross sections were processed in SCALE 4.2 with CSAS1X module which runs BONAMI-NITAWL-XSDRNPMS sequence. In this sequence XSDRNPMS does an infinite medium computation and computes the infinite multiplication factor. The 238-group ENDF/B-V cross sections were processed with CSAS1X. SCALE 4.3 has an improved resonance treatment in NITAWL that allows higher order resonance treatment. The 27-group cross sections and the 238-group in SCALE 4.2 employ only s-wave scattering but SCALE 4.3 allows p- and d-wave scattering (p-wave, $\ell = 2$ is the default value) with the 238-group cross sections. The ENDF/B-V and ENDF/B-VI cross sections are used in this study in MCNP. For MCNP the system was modeled as a large slab with reflecting boundary conditions. MCNP runs with ENDF/B-VI cross sections were run on workstations at Georgia Tech. A few cases run at Georgia Tech with ENDF/B-V cross sections repeated results in this study and confirmed that differences are cross section dependent and not machine dependent.

SAFE K VALUES

This study will establish safe-weight ratios for metal oxide poisons to U-235. The approach is to examine several cross sections and processing systems and to base the safe weight ratio on the cross section which computes the highest infinite multiplication factor. The difficult problem is to establish the calculated allowable maximum multiplication factor k_s. There are no critical experiments with iron, manganese, chromium or sodium with intermediate energy spectra. There have been measurements with highly enriched UO_2 fuel rods in dry and reflected lattices which have been used to establish bias values with SCALE 4.2 cross sections and KENO. These experiments could be used to provide guidance about bias for oxide systems but would be incomplete because of the metal poisons and because the experiments are not amenable for infinite medium calculations.

It is normal practice to determine a bias and bias uncertainty for a cross section set and processing system based on experimental data. It is not uncommon to then add an additional factor, sometimes called a minimum subcritical margin (MSM), to account for unknown and unquantified uncertainties. Because of the lack of experimental data, this study will consider several cross section sets and processing systems and establish a minimum safe weight ratio based on the highest computed (most conservative) infinite multiplication factor. A minimum subcritical margin of 0.05 ($k_s = 0.95$) will be used. The justifications for not including a bias and bias uncertainty are 1) the use of the highest computed multiplication factor with codes and cross sections available and 2) computation of the infinite multiplication factor which does not take into account neutron leakage. The two cross section systems, ENDF/B-V as processed with SCALE 4.3 238-group and MCNP with ENDF/B-V cross sections are in close agreement and are the basis for establishing the safe weight ratios for iron and sodium. For manganese and chromium MCNP with ENDF/B-VI cross sections produce higher multiplication factor than with the ENDF/B-V cross sections.

MATERIALS

The elements oxygen, sodium, chromium, manganese, iron and the isotope U-235 are included in this paper. The cross sections for these materials are summarized in Table 2. For this table the cross sections are taken from Mughabghab[4] et al. In this table are listed the thermal neutron absorption cross section, σ_γ, the scattering cross section, σ_s, the infinite dilute resonance integral and the approximate location of the first, or lowest energy, resonance. The latter quantity is listed to indicate that the poisons considered here all have resonances in the keV range and are significantly higher in energy than the resonances in the fissile material.

Table 2
Material Properties

Material	Prin. Iso. % abun.	σ_γ b	σ_s b	I_γ b	First Res. KeV
O	O-16 99.8%	0.19×10^{-3}	3.76	0.36×10^{-3}	430
Na	Na-23 100%	0.53	3.02	0.31	3
Cr	Cr-52 83.8%	3.07	3.38	1.6	5
Mn	Mn-55 100%	13.3	2.2	14.0	0.3
Fe	Fe-56 91.7%	2.59	12.46	1.4	1.15
U-235 (n,γ)	-	98.3	14.3	144	0.001
U-235 fission	-	582.6	14.3	275	0.001

IRON-URANIUM

Goslen demonstrated that a mixture of iron and U-235 with weight ratio 77:1 (atom ratio 324:1) is always safe. Goslen made mixtures $^{235}UO_2$ with density 10.96 g/cc and Fe_2O_3 with density 5.24 g/cc and modeled water as a solvent for the wet mixtures. Selected results from Goslen's study are in Table 3. In his study Goslen included a bias based on uranium aqueous data but did not include a bias for the absorber materials.

Table 3
K-infinite HRXN-ANISN
Fe:U-235 Atom Ratio = 324:1

H:U Atom Ratio	Calculated K	Bias Adjusted K
0.0	0.9443	0.949
10.27	0.9015	0.915
102.7	0.8721	0.902
205.5	0.8655	0.896
513.7	0.8269	0.851

These data indicate that small amounts of water add a strong negative reactivity.

The iron-uranium data from the Parks paper[3] are shown in Table 4.

Table 4
Reference 12 Computed K-Infinite Values
Fe:U Atom Ratio 320:1 No oxygen

Code and cross section	K-infinite
SCALE 218-gp ENDF/B-IV	0.8198
SCALE 238-gp ENDF/B-V	1.1446
Vitamin-E 174-gp ENDF/B-V	1.081
Vitamin-B6 199-gp ENDF/B-VI	1.1085
VIM continuous ENDF/B-V	1.0877±0.0007
MC^2 2040-gp ENDF/B-V	1.0965
MONK - UKNDL	1.0762±0.0012
MONK - JEF	0.9884±0.0008
MCNP continuous ENDF/B-V	1.0895±0.0017[a] 0.9901±0.0021[b]
MCNP continuous ENDF/B-VI	1.1135±0.0008

[a] Iron cross sections from 26000.50c from file endf5p
[b] Default iron cross section 26000.55c from file rmccs.

These computed k values spread from 0.82 to 1.14. Parks offers some reasons for the discrepancies and notes that the results for the SCALE-238 group (and 218-group) are high for two reasons:

1. The group structure is too broad in the region of some iron resonances that are important to this system.
2. The NITAWL procedure for resolved resonance processing calculates a background cross section using the scattering radius from ENDF. This assumption is inadequate for iron in the energy range of importance to this system.

The atom ratio Fe:U is close to that used by Goslen. The calculations reported in Table 4 do not include oxygen which is in Goslen's model. All but one of the data sets and processing codes in Table 4 predict a higher multiplication factor than computed by Goslen.

The iron-uranium system used by Parks et al. was analyzed with several cross section sets and processing codes available at SRS for both dry and wet systems. In these calculations the U-235 and iron atom densities were 2.6536E-04 and 8.4900e-02 atoms/b-cm respectively. Hydrogen was added to the mixture without including oxygen. For this table the SCALE 4.3 runs were not made on the RSK-6000 platform but with the PC version. The KENO and MCNP results are the computed multiplication values without correction for the statistical uncertainty. All Monte Carlo runs have a one standard deviation uncertainty of about 0.3%. The results are summarized in Table 5.

Table 5
Computed K Infinite Values
Fe:U = 320:1

	SCALE 4.3			SCALE 4.2	KENO	HRXN ANSN	MCNP	MCNP
H/U	27-gp	238-gp	44-gp	27-gp	27-gp	16-gp	Fe.50c	Fe-55c
0	0.8575	1.1466	1.1507	0.8352	0.8340	1.0028	1.0872	0.9983
10	0.8232	0.9713	-	0.8229	0.8340	0.9437	0.9579	0.9172
100	0.8444	0.8700	0.8712	0.8450	0.8454	0.8799	0.8651	0.8590
1000	0.7498	0.7542	0.7543	0.7498	0.7530	0.7682	-	-

These results only confirm the conclusion of Parks et al. that for dry systems there is a large range of computed multiplication factors with different cross section sets and processors. The need for experimental measurements is obvious. It is also noted that for a hydrogen-to-fissile ratio of 100 the spread in computed multiplication factors is reduced to about 3%.

The HRXN-ANISN runs were made with the same system used by Goslen, yet for the dry system the multiplication factor is about 15% higher than Goslen's results. Does oxygen make the difference? The 238-group library in SCALE 4.3 was used to answer two questions: what is the effect of including oxygen and what Fe:U ratio will bring the computed multiplication factor below 0.95. Two oxygen atoms were included for each uranium atom (UO_2) and 1.5 oxygen atom for each iron atom (Fe_2O_3). The results are in Table 6 for the 238-group cross sections for different Fe:U ratios and in Table 7 for various cross sections.

Table 6
SCALE 4.3 238-Group
K infinite

Fe/U	No Oxygen	With Oxygen
320	1.1465	0.9443
350	1.1068	0.9034
450	0.9919	0.7886
500	0.9426	0.7411
550	0.8980	0.6990

Table 7
Oxygen Effect
Fe:U = 320:1

	no oxygen	with oxygen
238-gp - SCALE 4.3	1.1465	0.9443
238-gp - SCALE 4.2	0.8602	0.8572
27-gp - SCALE 4.2	0.8352	0.8052
MCNP .50c	1.0879	0.9361
MCNP .55c	0.9987	0.8987
MCNP-ENDF/B-VI	1.1128	0.9368

These values indicate the importance of oxygen. The moderating effect of oxygen is such to shift the neutron spectrum toward the thermal region where the iron absorption becomes more important. KENO computations were made with the 27-group cross sections to find the average energy of neutrons causing fission. For the uranium-iron mixture without oxygen, the average energy of neutrons causing fission is 14.8 keV. With oxygen in the system the value drops to 34 eV. With the addition of water at H/U = 100 the value is 0.5 eV. These values indicate that the system is not well thermalized until about H/U=100. This is further illustrated in Figure A-1 in the appendix which shows the energy-dependent fission rate.

In Table 7 there is a significant difference in the 238-group results between SCALE 4.2 and SCALE 4.3. The scattering option can be changed in SCALE 4.3 with the "more data" option ("more data mlv=1 end more data" will call p-wave scattering). For s-wave scattering ($\ell = 0$) with oxygen in the system the computed k value is 0.8554, which is close to the SCALE 4.2 value. For p-wave scattering ($\ell = 1$) the value is 0.9407, close to the SCALE 4.3 value with the default value, $\ell = 2$. For this Fe-U mixture with water at H/U =100, the K values are 0.8635 for $\ell = 2$ and 0.8515 for $\ell = 0$, not much difference. This result means that the inclusion of the higher order scattering terms affects the computed multiplication factor where resonance scattering by heavier-than-hydrogen materials is important.

The infinite multiplication factors for mixtures with oxygen as computed with the 238-group cross sections in SCALE 4.3 and with MCNP with both ENDF/B-V and ENDF/B-VI cross sections agree within 1%. With oxygen in the system, the 238-group and 27-group cross sections processed with CSAS1X in SCALE 4.2 and 4.3 as well as both Fe cross section sets in MCNP predict the infinite multiplication factor less than 0.95 for an iron-to-uranium atom ratio of 320. This defines a safe atom ratio of Fe:U-235 of 320:1 (weight ratio 78:1).

MANGANESE-URANIUM

Goslen demonstrated that a mixture of manganese and U-235 with Mn:U-235 weight ratio greater than 30:1 (atom ratio 128:1) is always safe. Some of Goslen's results are in Table

8. Goslen made mixtures of UO_2 with density 10.96 g/cc and MnO_2 with density 5.026 g/cc and modeled water as a solvent for the wet mixtures.

Table 8
HRXN Computed K-Infinite[1]
Mn:U weight ratio 30:1

H:U Atom Ratio	Calculated K	Bias Adjusted K
0.0	0.9310	0.936
14.9	0.7102	0.726
104.1	0.5850	0.615
223.2	0.5700	0.600

As with the iron-uranium mixtures water adds a strong negative reactivity.

Mixtures of U-235 and manganese were modeled with and without oxygen at a Mn:U-235 weight ratio 30:1 (atom ratio 128:1). The atomic densities for U-235 and manganese were 2.6831E-04 and 3.4437E-02 atoms/b-cm respectively. Oxygen was included as two oxygen atoms for each atom of uranium (UO_2) and two for each atom of manganese (MnO_2). The computed multiplication factors are in Table 9. The ENDF/B-V manganese 25055.50c was used in the MCNP calculations. The MCNP results are the computed multiplication values without correction for the statistical uncertainty. They have a one standard deviation uncertainty of about 0.2%.

Table 9
K-Infinite Mn:U-235 Mixtures
Atom Ratio 128:1

	No Oxygen	With Oxygen
238-Gp SCALE 4.3	1.1713	0.5924
238-Gp SCALE 4.2	0.8423	0.5955
27-Group	0.7395	0.5165
MCNP ENDF/B-V	0.9194	0.5996

As with the iron-uranium mixtures, the oxygen sufficiently moderates the spectrum to drive the multiplication factor down. Manganese with its large thermal neutron absorption cross section and large resonance integral, 13.3 and 14.0 barns respectively, is an effective poison and these results indicate that Goslen's results with the Hansen Roach cross section processed with HRXN are conservative. As an example, the 238-group cross section set in SCALE 4.3 were used with varying Mn:U ratios. The results are in Table 10.

Table 10
K-Infinite Mn:U-235 Mixtures
238-Group Cross Sections

Atom Ratio Mn:U	Atom Ratio H:U=0	Atom Ratio H:U=10	Atom Ratio H:U=100
50	0.9810	-	-
54	0.9464	0.8613	0.8783
60	0.9004	0.8241	0.8292
65	0.8657		
70	0.8340	-	-

These data indicate that in the limited range of the Mn:U-235 atom ratios from 50 to 70 there is a strong dependence of the multiplication factor on the atom ratio. For a Mn:U-235 atom ratio of 54:1 (weight ratio 13:1), the infinite multiplication factor is less than 0.95 with the 238-group cross sections and the addition of hydrogen to this mixture decreases the multiplication factor. There is good agreement between SCALE 4.3 with 238-group cross sections and ENDF/B-V cross section with MCNP (k = 0.9546), however, at this ratio the ENDF/B-VI cross sections compute a multiplication factor of 1.02. A search for a safe ratio with the ENDF/B-VI cross sections was made. Multiplication factors for a Mn:U-235 atomic ratio 65:1 are summarized in Table 11.

Table 11
K-Infinite Mn:U-235 Mixtures
Atom Ratio 65:1

	With Oxygen
238-Gp SCALE 4.3	0.8657
238-Gp SCALE 4.2	0.8320
27-Group	0.8005
MCNP-ENDF/BV	0.8780
MCNP-ENDF/BVI	0.9357

The MCNP results are the computed multiplication values without correction for the statistical uncertainty. They have a one standard deviation uncertainty of about 0.3%. With oxygen in the system the 238-group and 27-group cross sections processed with CSAS1X in SCALE 4.2 and 4.3 and MCNP with both the ENDF/B-V and ENDF/B-VI cross sections predict the infinite multiplication factor less than 0.95 for a manganese-to-uranium atom ratio of 65. These results define a safe atom ratio of Mn:U-235 of 65:1.

CHROMIUM-URANIUM

Goslen demonstrated that a mixture of chromium and U-235 with Cr:U-235 weight ratio greater than 52:1 (atom ratio 235:1) is always safe. Some of Goslen's results are in Table

12. Goslen made mixtures of UO_2 with density 10.96 g/cc and Cr_2O_3 with density 5.216 g/cc and modeled water as a solvent for the wet mixtures.

Table 12
HRXN Computed K-Infinite
Cr:U-235 Atom ratio 235:1

H:U Atom Ratio	Calculated K	Bias Adjusted K
0.0	0.7847	0.790
12.3	0.8266	0.841
91.95	0.9084	0.938
122.6	0.9155	0.946
214.5	0.9185	0.949
919.5	0.8207	0.830

For Cr:U-235 mixtures with HRXN the most reactive combination is not the dry system.

Mixtures of U-235 and chromium were modeled with and without oxygen at a Cr:U-235 weight ratio 52:1 (atom ratio 235:1). The atomic densities for U-235 and chromium were 1.7460E-04 and 4.1041E-02 atoms/b-cm respectively. Oxygen was included as two oxygen atoms for each atom of uranium (UO_2) and 1.5 for each atom of chromium (Cr_2O_3). The computed multiplication factors are in Table 13. The recommended ENDF/B-V chromium 24000.50c was used in MCNP. The MCNP results are the computed multiplication values without correction for the statistical uncertainty. They have a one standard deviation uncertainty of about 0.3%.

Table 13
K-Infinite Values
Cr:U-235 Atom Ratio 235:1

	SCALE 4.2 27-group		SCALE 4.3 238-group		MCNP ENDF/BV	
H/U	no oxy	with oxy	no oxy	with oxy	no oxy	with oxy
0	0.7941	0.9006	0.9451	0.9675	0.8857	0.9639
1	0.8298	0.8983	0.9816	0.9583	0.9383	0.9535
10	0.8817	0.8859	0.9439	0.9161	0.9317	0.9079
100	0.9042	0.9025	0.9079	0.9050	0.9066	0.9006
1000	0.7888	0.7877	0.7906	0.7892	0.7923	0.7931

For each cross section set oxygen increases the multiplication factor for the dry case and for each cross section set with oxygen the reactivity decreases as water is added. This is different from the results with the Hansen Roach cross sections. It is also noted that the 238-group cross sections in SCALE 4.3 agrees well with MCNP with ENDF/B-V cross sections. The data suggest that a Cr:U-235 atom ratio 235:1 may not be conservative. Increasing the Cr:U ratio will decrease the multiplication factor. Table 14 shows this trend as computed with the 238-group cross sections, and Table 15 includes the results for several cross section sets.

Table 14
K Infinite
Dry Cr:U-235 with Oxygen
Scale 4.3 238-Group Cross Sections

Cr:U Atom Ratio	k
250	0.9423
265	0.9182
280	0.8952
290	0.8804

Table 15
K-Infinite Cr:U-235 Mixtures
Atom Ratio 290:1

	With Oxygen
238-Gp SCALE 4.3	0.8804
238-Gp SCALE 4.2	0.8206
27-Group	0.8196
MCNP-ENDF/BV	0.8789
MCNP-ENDF/BVI	0.9523

MCNP with ENDF/B-VI cross sections compute the highest infinite multiplication factor. The multiplication factors for Cr-U-235 mixtures with oxygen as computed with the 238-group cross sections in SCALE 4.3 and with MCNP with ENDF/B-V cross sections agree within 1.0%. With oxygen in the system the 238-group and 27-group cross sections processed with CSAS1X in SCALE 4.2 and 4.3 and MCNP with ENDF/B-V cross sections predict the infinite multiplication factor less than 0.95 for a chromium-to-uranium atom ratio of 290. However, the MCNP with ENDF/B-VI cross sections exceeds 0.95 for a Cr:U-235 290:1 atom ratio. The ENDF/B-VI cross sections give a multiplication factor 0.937 for a Cr:U-235 300:1 atom ratio. This result defines a safe atom ratio of Cr:U-235 of 300:1.

SODIUM-URANIUM

Sodium uranium systems were not studied by Goslen and it is recognized that sodium might not be a particularly effective poison because of the low cross section. However, sodium is a major constituent in some waste streams. For example the composition of samples from saltcake in Tank 41H has been measuerd. These measurements indicate that both the soluble and insoluble components of the salt contain 10 to 20 weight percent sodium. In all cases the sodium-to-fissile atom ratio exceeds 3000.

A mixture of U-235 and sodium was modeled at an atom ratio Na:U-235 of 1000:1 by mixing UO_2 (density 10.46 g/cc) and $NaNO_3$ (density 2.261 g/cc). The atom densities for U-235 and Na in this mixture are 1.6009E-05 and 1.6009E-02 atoms/b-cm respectively. Sodium may exist in waste in several forms, for example as nitrate, carbonate hydroxide or oxide, each of which has a different oxygen content. For this study one oxygen atom was included with each sodium atom and two oxygen atoms with each uranium atom. A search with the 238-group cross sections in SCALE 4.3 showed that for a Na:U-235 atom ratio 1300:1 (weight ratio 127:1) the infinite multiplication factor is less than 0.95 for the dry system with oxygen. Computations on this system are summarized in Table 16 for the three cross section sets. The ENDF/B-V sodium cross section 11023.50c was used in MCNP.

Table 16
K Infinite for Sodium-Uranium Mixture
Na:U-235 = 1300:1

H/U	SCALE 4.3 238 gp	SCALE 4.2 238 gp	SCALE 4.2 27 gp	MCNP ENDF/B-V	MCNP ENDF/B-VI
0	0.9484	0.9471	0.9082	0.9551	0.9423
1	0.9483	0.9470	0.9090	0.9518	n.a.
10	0.9482	0.9471	0.9150	0.9501	n.a.
100	0.9482	0.9477	0.9335	0.9489	n.a.
1000	0.8087	0.8089	0.8023	0.8067	n.a.
0 (no oxygen)	1.0111	1.0028	0.8444	1.0082	0.9907

The H/U ratio is an atom ratio and all of the MCNP runs have a one standard deviation uncertainty less than 0.3%. Here there is good agreement between the 238-group cross sections and MCNP with both cross section sets. In this analysis oxygen and sodium are both present at more than 1000 times the U-235 content. Thus the addition of water to the mixture does not make much difference until a significant amount of hydrogen is added. The MCNP case with ENDF/B-V cross sections computes an infinite multiplication factor slightly greater than 0.95. Increasing the Na:U-235 ratio to 1325 reduces the computed multiplication factor to 0.9435. This result defines a safe atom ratio of Na:U-235 of 1325:1.

CONCLUSION AND SUMMARY

Several investigators have shown that there may be a wide range of computed multiplication factors with different cross section sets and cross section processing codes in mixtures of fissile materials and metals in which the neutron spectrum is not well thermalized. This observation raised the question of the validity of using poisons as criticality controls in waste materials. This could be of a particular concern if a waste stream with water has the possibility of drying out.

Infinite multiplication factors have been computed for mixtures of U-235 with iron, manganese, chromium and sodium with several cross section sets. Computations were done for metal mixtures, mixtures of metal oxides, and with admixtures of water. The computation tools used are the 27-group cross sections in SCALE 4.2 processed with the CSAS1X module, the 238-group cross sections in SCALE 4.2 and 4.3 processed with CSAS1X and MCNP 4.a with ENDF/B-V and ENDF/B-VI cross sections. For water-moderated and oxide systems there was good agreement between the 238-group cross sections and MCNP with ENDF/B-V cross sections. The 27-group cross section set in general calculated lower multiplication factors. For manganese and chromium the ENDF/B-VI cross sections predict higher multiplication factors than ENDF/B-V cross sections.

The oxygen in the oxide systems provides sufficient moderation to drive the neutron spectrum in these metal systems towards the thermal region. In these more thermal systems the spread in computed multiplication factors among different cross sections sets is reduced. Many of the waste streams contain oxides and a premise of the study is that oxygen will always be present. Maximum infinite multiplication factors and metal to U-235 ratios are summarized in Table 17.

Table 17
Maximum K Infinite Values
Metal Oxide Mixtures

Mixture	Weight Ratio	Atom ratio	K infinite
Fe:U-235	76:1	320:1	0.95
Mn:U-235	15:1	65:1	0.95
Cr:U-235	66:1	300:1	0.95
Na:U-235	130:1	1325:1	0.95

ACKNOWLEDGMENT

MCNP runs with ENDF/B-VI cross sections were run at Georgia Tech on workstations in the Nuclear Engineering and Health Physics Program. The assistance of Michelle Pitts and her adviser, Dr. Farzad Rahnema, is gratefully acknowledged.

REFERENCES

1. B. Palmer "K∞ for Certain Metals Mixed With ^{235}U", *Criticality Safety Quarterly*, Winter 1993.

2. C. V. Parks et al., "Oak Ridge Study Reveals Uncertainty in K∞ Values for Certain Metals Mixed with ^{235}U", *Criticality Safety Quarterly*, Fall 1994.

3. C. V. Parks, W. C. Jordan, L. M. Petrie and R. Q. Wright, "Use of Metal/Uranium Mixtures to Explore Data Uncertainties", TANSAO 73, 217, 1995.

4. S. F. Mughabghab, M. Divadeenam and N. E. Holden, Neutron Cross Sections, Vol. 1, Academic Press, New York, 1981.

APPENDIX A: ADDED INFORMATION

Several additional pieces of information computed by the codes are included in the appendix for the interested reader. The code XSDRNPM computes the energy dependent fission, absorption and leakage rate. Plots of the fission rate with the 27-group cross sections are shown to illustrate the change in spectrum with changing conditions. Also listed are some of the values of the average energy of neutrons causing fission as computed with KENO for the 27-group cross sections and the effect of changing the scattering option in SCALE 4.3 with the 238-group cross sections.

IRON-U-235

Figure A.1
Fe:U-235 320:1

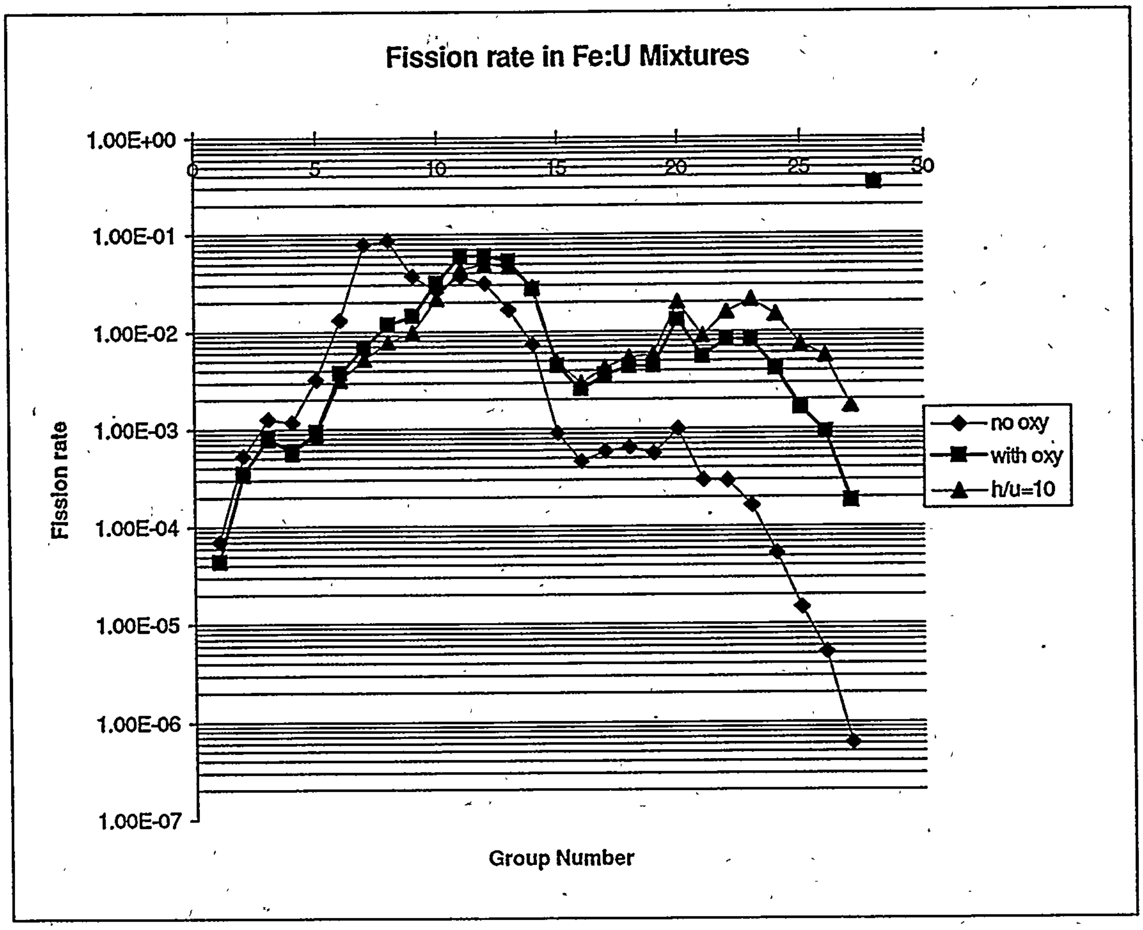


Figure A.1 shows the fission rate for the 27-group cross sections for an Fe-U-235 atom ratio 32:1. The computed k_{inf} are 0.8352 for the mixture with no oxygen, 0.8052 with oxygen but no hydrogen and 0.8126 for the H/U = 10 case. As noted in Table 7 the 238-group cross sections in SCALE 4.3 give a much larger difference between the two cases with and without oxygen. Figure A.1 shows a marked difference in the fission rates for groups higher than group 15 with the inclusion of oxygen and water in the system. Group 15 has an upper energy of 3.05 eV (see Table A.1). With hydrogen the fission rate in groups around group 23 is further enhanced.

Table A.1
27-Group Energy Structure

Group	Upper Lethargy	Upper Energy MeV	Group	Upper Lethargy	Upper Energy keV
1	-0.693	20	6	2.408	900
2	0.441	6.43	7	3.219	400
3	1.204	3	8	4.605	100
4	1.687	1.85	9	6.377	17
5	1.966	1.4	10	8.112	3

Group	Upper Lethargy	Upper Energy eV	Group	Upper Lethargy	Upper Energy eV
11	9.808	550	20	16.341	0.8
12	11.503	100	21	17.034	0.4
13	12.717	30	22	17.242	0.325
14	13.816	10	23	17.61	0.225
15	15.03	3.05	24	18.421	0.1
16	15.547	1.77	25	19.114	0.05
17	15.856	1.3	26	19.625	0.03
18	15.996	1.13	27	20.723	0.001
19	16.118	1	Bottom		1.00E-05

KENO calculations which compute the average energy of neutrons causing fission are summarized in Table A.2. For these cases the KENO model was a large slab with reflecting boundary conditions and was run until the one standard deviation uncertainty was less than 0.2%. Results from similar XSDRN runs are shown for comparison.

Table A.2
Average Energy Causing Fission
27-Group

Fe/U=320	k_{eff} XSDRN	k_{eff} KENO	27-Group	Energy
no oxygen	0.8352	0.8340	9.08	14.8 kev
no oxy H/U=1	n.a.	0.8298	9.82	4.1 kev
with oxy.	0.8052	0.8053	12.9	34 ev

These values further illustrate the fission rate densities shown in Figure A.1.

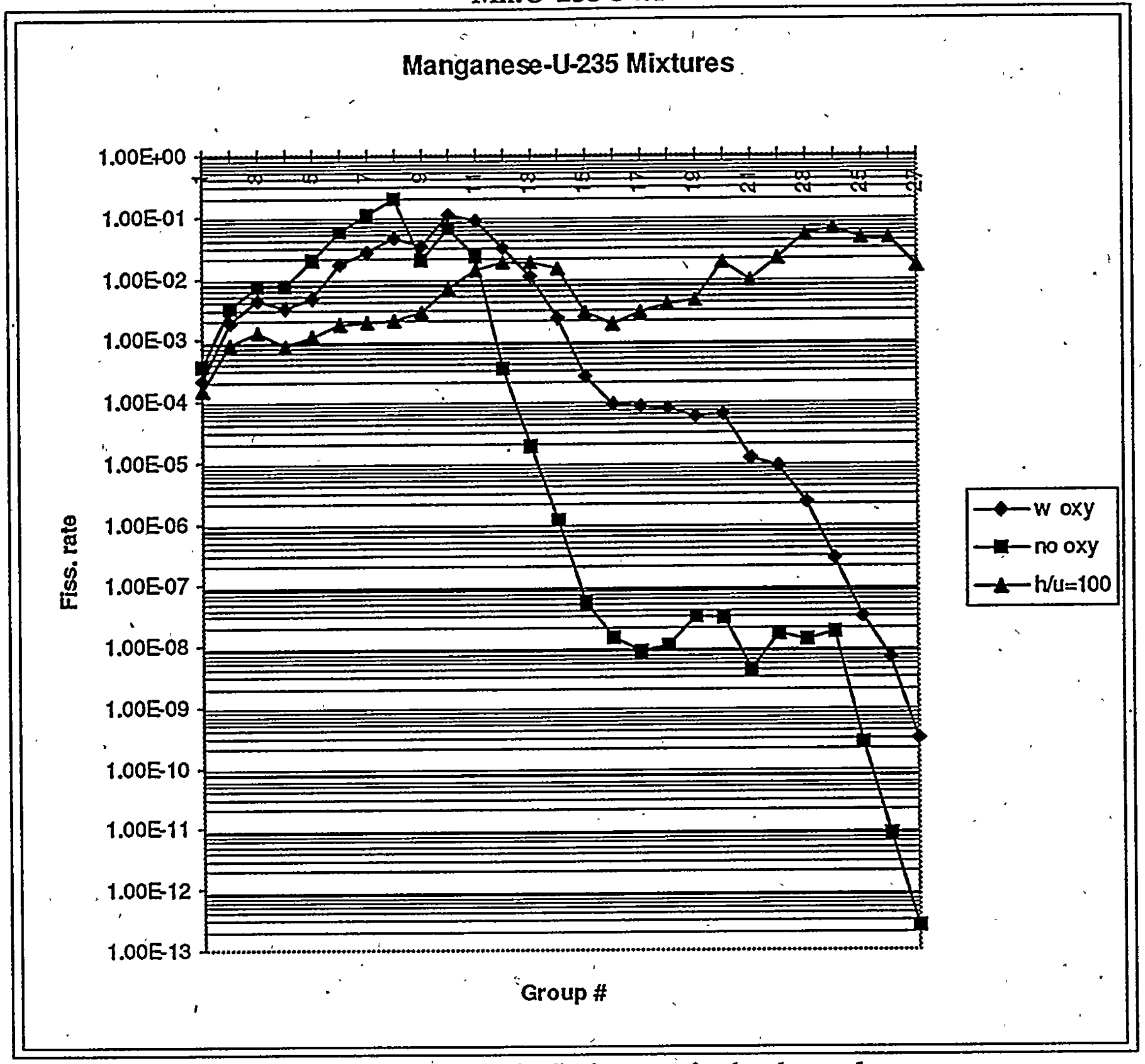
The effect of scattering order is included in Table A.3. The scattering option can be changed in SCALE 4.3 with the "more data" option ("more data mlv=1 end more data" will call p-wave scattering). The default value is $\ell=2$, d-wave scattering. SCALE 4.2 does not have this option and allows only s-wave, $\ell=0$, scattering. The columns headed H/U=1 and H/U=10 have no oxygen.

Table A.3
Infinite Multiplication Factors
Effect of Scattering Order
SCALE 4.3 238-group

Fe/U=320	H/U=1	H/U=10	H/U=0 With Oxy	No Oxy
l=2	1.1042	0.9713	0.9443	1.1465
l=1	1.0566	0.9597	0.9407	1.0708
l=0	0.8766	0.8718	0.9554	0.8749

MANGANESE-U-235

Figure A.2
Mn:U-235 54:1



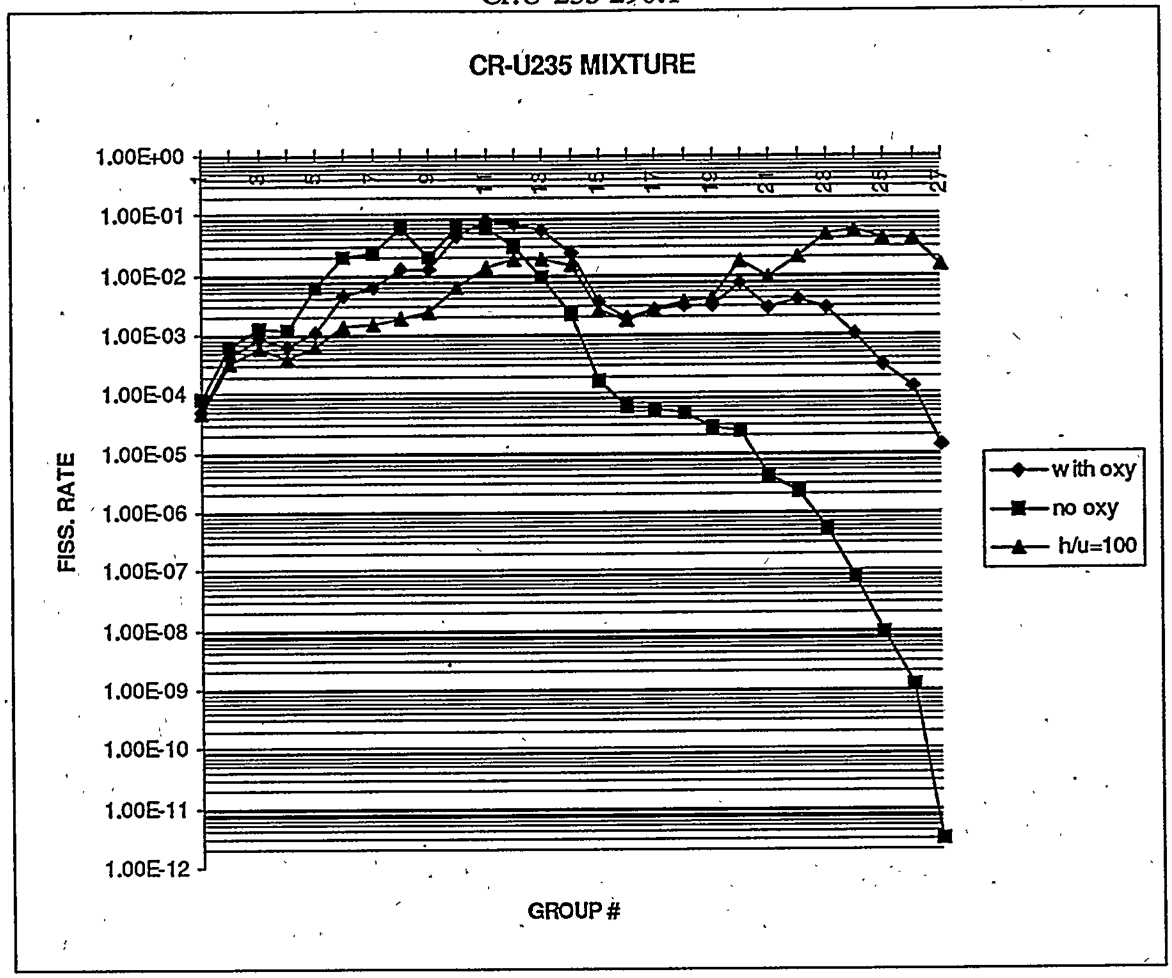
This figure illustrates a dramatic change in fission rate in the thermal neutron energy range as first oxygen and then hydrogen are included in the system. The inclusion of oxygen increases the fission rate in groups with group numbers higher than 25 by about three orders of magnitude, and adding water further increases the fission rate. This phenomena is further reflected in the change in the average energy causing fission shown in Table A.4.

Table A.4
Average Energy Causing Fission
27-group

Mn/U=54	K-KENO	27-Group	Energy
no oxygen	1.2118	7.70	151 kev
with oxy.	0.8883	9.56	6.43 kev
no oxy H/U=100	0.8707	20.82	0.45 ev

CHROMIUM URANIUM

Figure A.3
Cr:U-235 290:1



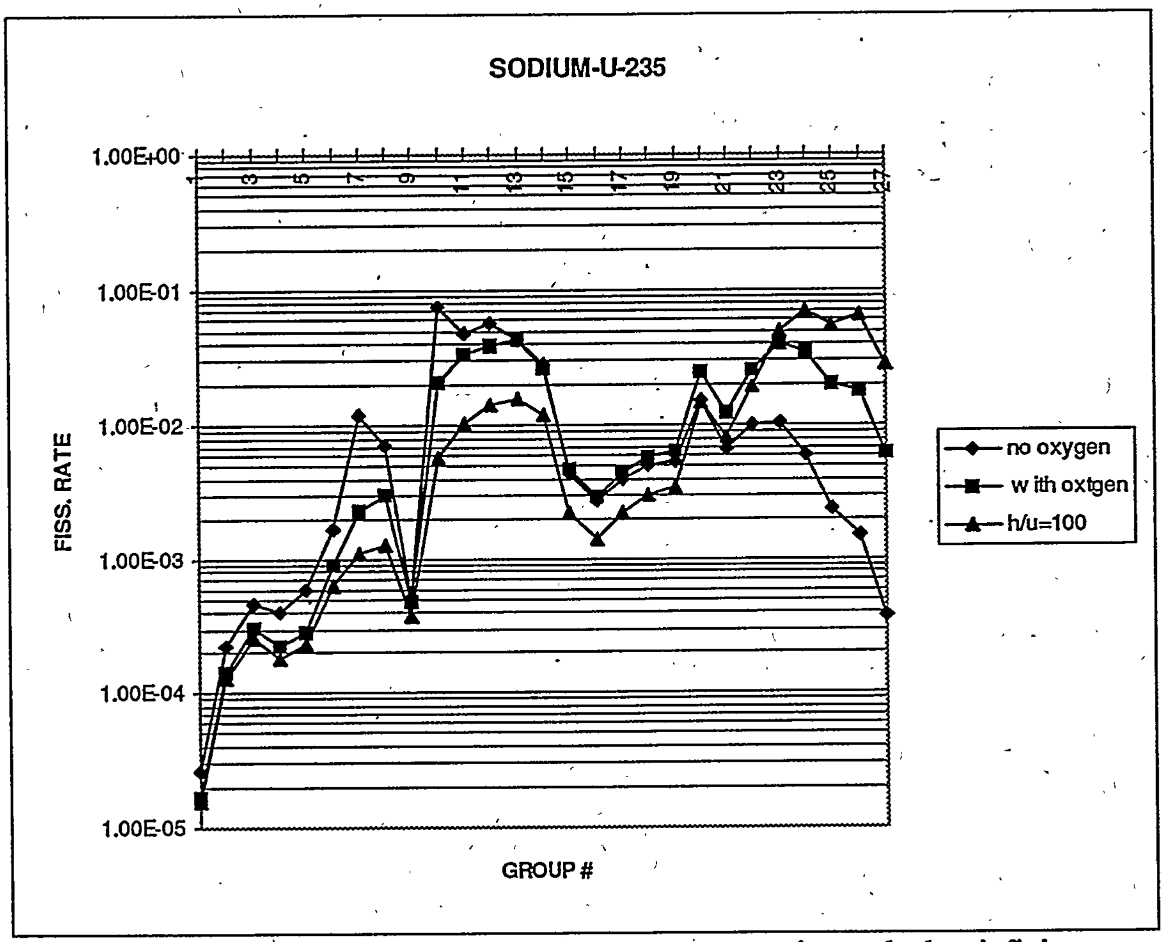
As with the others this figure illustrates the increased thermalization as first oxygen and the water is included in the mixture. This is further reinforced by the average energy of neutrons causing fission as is listed in table A. 5.

Table A.5
Average Energy Causing Fission
27-group

Cr/U=290	K-KENO	27-Group	Energy
no oxygen	0.7159	9.36	91 kev
with oxy.	0.8228	12.02	98.6 ev
no oxy H/U=100	0.8039	20.78	0.47 ev

SODIUM-URANIUM

Figure A.4
Na:U-235 1300:1



For the cases with Na:U-235 = 1300:1 the 27-group cross sections calculate infinite multiplication factors of 0.8444 with no oxygen in the system, 0.9082 with the inclusion of oxygen and 0.9489 for H/U=100. The fission rate spectra indicate that this amount of sodium fairly well thermalizes the spectrum but the presence of oxygen and hydrogen further enhances the thermal fission.

APPENDIX B: SAMPLE INPUT FILES

1.0) 238-group, fe/u=320
with oxygen

```
=csas1x   parm=size=600000
fe/u problem fe/u=320
238groupndf5 infhommedium
u-235  1  0.0  2.6536e-04  298  end
fe     1  0.0  0.0849  298  end
o      1  0.0  0.1279  298  end
end comp
end
```

2.0) 27-group fe/u= 320
h/u=1 no oxygen

```
=csas1x  parm=sixe=600000
fe/u problem h/u = 1
27groupndf4 infhommedium
u-235  1  0.0  2.6536e-04  298  end
fe     1  0.0  0.0849  298  end
h      1  0.0  2.6536e-04  298  end
end comp
end
```

3.0) MCNP fe/u=320
with oxygen

```
fe/u infinite medium with oxy
c    cell cards
1    1 2.1305e-1 -1 2 -3 4 -5 6 imp:n=1
2    0          1 -2 3 -4 5 -6 imp:n=0

c surface cards
*1  px  20
*2  px  -20
*3  py  100
*4  py  -100
*5  pz  100
*6  pz  -100

c data cards
m1  92235.50c  2.6536e-4
    26000.55c  8.4900e-2
    8016.50c   1.2788e-1
kcode 100 1.0 15 115
ksrc  0 0 0
```

4.0) 27-group mn/u=54
with oxygen

```
=csas1x    parm=size=600000
mn/u problem mn/u=54
27groupndf4 infhommedium
u-235  1  0.0  2.6831e-04  298  end
mn     1  0.0  1.4488e-02  298  end
o      1  0.0  2.9514e-02  298  end
end comp
end
```

5.0) 238-group mn/u=60
h/u=10

```
=csas1x    parm=size=600000
mn/u  mn/u=60 h/u-10
238groupndf5 infhommedium
u-235  1  0.0  2.6831e-04  298  end
mn     1  0.0  1.6098e-02  298  end
o      1  0.0  3.2733e-02  298  end
h      1  0.0  2.6831e-03  298  end
end comp
end
```

6.0) 27-group keno
mn/u=54, with oxygen

```
=csas25  parm=sixe=600000
mn/u problem keno h/x=0
27groupndf4 infhommedium
u-235  1  0.0  2.6831e-04  298  end
mn     1  0.0  1.4488e-02  298  end
o      1  0.0  2.9514e-02  298  end
end comp
fe/u problem h/x=0
read param
tme=90 nub=yes
end param
read geometry
cuboid 1 1 100 -100 100 -100 100 -100
end geom
read bnds
xfc=mirr yfc=mirr zfc=mirr
end bnds
end data
end
```

7.0) MCNP mn/u= 65
with oxygen

```
mn/u infinite medium  mn/u=65
c    cell cards
1    1 5.3125e-02  -1 2 -3 4 -5 6 imp:n=1
2    0          1 -2 3 -4 5 -6 imp:n=0

c surface cards
*1  px  20
*2  px  -20
*3  py  100
*4  py  -100
*5  pz  100
*6  pz  -100

c data cards
m1   92235.50c  2.6831e-4
     25055.50c  1.7440e-2
     8016.50c   3.5416e-2
kcode 100 1.0 15 115
ksrc  0 0 0
```

8.0) 27-group cr/u=290
no oxygen

```
=csas1x   parm=size=600000
cr/u problem cr/u=290
27groupndf4 infhommedium
u-235  1  0.0  1.7460e-04  298  end
cr     1  0.0  5.0633e-02  298  end
end comp
end
```

9.0) 238-group cr/u=235
with oxygen

```
=csas1x   parm=size=600000
cr/u problem  h/u=1
238groupndf5 infhommedium
u-235  1  0.0  1.7460e-04  298  end
cr     1  0.0  4.1041e-02  298  end
h      1  0.0  1.7460e-04  298  end
o      1  0.0  6.1998e-02  298  end
end comp
end
```

10) MCNP cr/u=290
with oxygen

```
cr/u infinite medium
c    cell cards cr/u=290
1    1 1.2711e-1  -1 2 -3 4 -5 6 imp:n=1
2    0          1 -2 3 -4 5 -6 imp:n=0

c surface cards
*1  px  20
*2  px  -20
*3  py  100
*4  py  -100
*5  pz  100
*6  pz  -100

c data cards
m1   92235.50c  1.7460e-4
     24000.50c  5.0633e-2
     8016.50c   7.6299e-2
kcode 100 1.0 15 115
ksrc  0 0 0
```

11) 27-group na/u=1300
with oxygen, h/u=1

```
#csas1x   parm=yes
u/na u/na=1300 h/u=1
27groupndf4        infhommedium
u-235 1 0.0 1.6009e-05 298 end
na    1 0.0 2.0812e-02 298 end
o     1 0.0 2.0852e-02 298 end
h     1 0.0 1.6009e-05 298 end
end comp
end
```

12) MCNP na/u=1300
with oxygen

```
na/u infinite medium  na/u=1300
c    cell cards
1    1 4.1656e-2  -1 2 -3 4 -5 6 imp:n=1
2    0        1 -2 3 -4 5 -6 imp:n=0

c surface cards
*1  px  20
*2  px  -20
*3  py  100
*4  py  -100
*5  pz  100
*6  pz  -100

c data cards
m1   92235.50c  1.6009e-5
     11023.50c  2.0812e-2
     8016.50c   2.0844e-2
kcode 100 1.0 15 115
ksrc  0 0 0
```