Metal Poisons For Criticality in Waste Streams (U)

by

T. G. Williamson
Westinghouse Savannah River Company
Savannah River Site
Aiken, South Carolina 29808
A. Q. Goslen

19980407 012

A document prepared for AMERICAN NUCLEAR SOCIETY

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 Rd., Springfield, VA 22161
INTRODUCTION

Many of the wastes from processing fissile materials contain metals which may serve as nuclear criticality poisons. It would be advantageous to the criticality evaluation of these wastes to demonstrate that the poisons remain with the fissile materials and to demonstrate an always safe poison-to-fissile ratio. The first task, demonstrating that the materials stay together, is the job of the chemist, the second, calculating an always safe ratio, is an object of this paper.

In an earlier study we demonstrated safe ratios for Fe, Mn and Cr oxides to $^{235}$U. In these studies the Hansen Roach 16-group cross sections were used with the SRS code HRXN. Multiplication factors were computed and safe ratios were defined such that the bias adjusted $k$ values were less than 0.95. These safe weight ratios are Fe:$^{235}$U - 77:1; Mn:$^{235}$U - 30:1 and Cr:$^{235}$U - 52:1.

Palmer $^1$ has shown that for certain mixtures of Al, Fe and Zr with $^{235}$U the computed infinite multiplication factors may differ by as much as 20% with different cross sections and processing systems. Parks et al.$^{23}$ have further studied these mixtures 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.

The purpose of this study is to reexamine our earlier work with cross sections and processing codes used at WSRC today. This study will focus on $^{235}$U mixtures with Fe, Mn, and Cr. Sodium will included in the list of poisons since sodium is an abundant element in many of the waste materials.

DISCUSSION

Computations were done on the RSK-6000 workstation cluster with Scale 4.2, Scale 4.3 and MCNP 4a. The 27-group cross sections were processed in Scale 4.2 with CSAS1X module which runs BONAMI-NITAWL-XSDRNPMS and computes the infinite multiplication factor. The 238-group cross sections were processed with CSAS1X in Scale 4.3 because of the improved resonance treatment in NITAWL that allows treatment of higher order scattering. In MCNP the ENDF/B-V (.50c) cross sections were used and the system was modeled as a slab with reflecting boundary conditions. All cases were run to a one standard deviation uncertainty of less than 0.3%.

The iron-uranium mixture investigated by Parks et al. has an Fe:U atom ratio of 320:1 and does not include oxygen. Our mixture had an Fe:U ratio 324:1 and included both iron and uranium as oxides. For this study mixtures were made with $^{235}$U and Fe atom densities of 2.6536E-04 and 8.49e-02 respectively. For the oxides two oxygen atoms were included for each uranium atom ($\text{UO}_2$) and 1.5 oxygen atom for each iron atom ($\text{Fe}_2\text{O}_3$).
Manganese mixtures with weight ratio Mn:235U of 30:1 (atom ratio 128:1) were made with atomic densities for 235U and Mn of 2.683E-04 and 3.443E-02 respectively. Two oxygen atoms were included for each manganese atom (MnO2). The chromium mixture has a Cr:235U weight ratio 52:1 (atom ratio 235:1) with atomic densities for 235U and Cr of 1.7460E-04 and 4.1041E-02 respectively. Two oxygen atoms were included for each uranium atom and 1.5 for each chromium atom (Cr2O3). A sodium mixture was made with an atom ratio Na:235U of 1450:1 with 235U and Na atom densities of 1.6009E-05 and 2.3213E-03 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. Infinite multiplication factors for these mixtures are in Table 1.

Table 1
Infinite Multiplication Factors

<table>
<thead>
<tr>
<th>Mixture Atm. ratio</th>
<th>Cross section</th>
<th>K-inf. Metal</th>
<th>K-inf. Oxide</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe:U 320:1</td>
<td>27-group</td>
<td>0.8352</td>
<td>0.8052</td>
</tr>
<tr>
<td></td>
<td>238-group</td>
<td>1.1465</td>
<td>0.9443</td>
</tr>
<tr>
<td></td>
<td>MCNP</td>
<td>1.0851</td>
<td>0.9346</td>
</tr>
<tr>
<td>Mn:U 128:1</td>
<td>27-group</td>
<td>0.7395</td>
<td>0.5165</td>
</tr>
<tr>
<td></td>
<td>238-group</td>
<td>1.1713</td>
<td>0.5924</td>
</tr>
<tr>
<td></td>
<td>MCNP</td>
<td>0.9194</td>
<td>0.5996</td>
</tr>
<tr>
<td>Cr:U 235:1</td>
<td>27-group</td>
<td>0.7941</td>
<td>0.9006</td>
</tr>
<tr>
<td></td>
<td>238-group</td>
<td>0.9451</td>
<td>0.9675</td>
</tr>
<tr>
<td></td>
<td>MCNP</td>
<td>0.8857</td>
<td>0.9639</td>
</tr>
<tr>
<td>Na:U 1450:1</td>
<td>27-group</td>
<td>0.8013</td>
<td>0.8619</td>
</tr>
<tr>
<td></td>
<td>238-group</td>
<td>0.9620</td>
<td>0.8997</td>
</tr>
<tr>
<td></td>
<td>MCNP</td>
<td>0.9570</td>
<td>0.9051</td>
</tr>
</tbody>
</table>
This table illustrates the point that there can be a wide range of computed multiplication factors for metal systems in which the neutrons are not well thermalized. It also illustrates the fact that oxygen is a reasonable moderator and most of the variability is removed with the oxides. For oxides there is good agreement between the 238-group computations and MCNP.

The iron results agree with our earlier study in which a computed $k$ of 0.95 defined the safe ratio. The manganese results are quite different from the earlier study and suggest that a lower manganese ratio might be appropriate. The 238-group cross sections calculate a Mn:$^{235}\text{U}$ safe weight atom ratio of 54:1 for infinite multiplication factor of 0.95. The chromium results also differ from the earlier study. In the earlier study the highest $k$ value occurred in a wet system with an H/U ratio of about 200. In this study the highest $k$ value is for the dry system and is higher than the safe value 0.94. The 238-group cross sections calculate a Cr:$^{235}\text{U}$ safe weight atom ratio of 250:1 for infinite multiplication factor of 0.95.

CONCLUSION

Safe weight ratios have been computed for mixtures of $^{235}\text{U}$ with iron, manganese, chromium and sodium. These materials often exist in waste materials in chemical forms with oxygen. The oxygen provides sufficient moderation that in dry systems the neutrons are reasonably thermalized. These results further reinforce the need for additional experimental data for these mixtures.
REFERENCES


