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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.[2,3] 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 (UO_2) and 1.5 oxygen atom for each iron atom (Fe_2O_3).

Manganese mixtures with weight ratio Mn:^{235}U of 30:1 (atom ratio 128:1) were made with atomic densities for ^{235}U and Mn of 2.6831E-04 and 3.4437E-02 respectively. Two oxygen atoms were included for each manganese atom (MnO_2). The chromium mixture has a Cr:^{235}U weight ratio 52:1 (atom ratio 235:1) with atomic densities for ^{235}U 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 ($Cr_2 O_3$). A sodium mixture was made with an atom ratio Na:^{235}U of 1450:1 with ^{235}U 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

Mixture Atm. ratio	Cross section	K-inf. Metal	K-inf. Oxide
Fe:U	27-group	0.8352	0.8052
320:1	238-group	1.1465	0.9443
	MCNP	1.0851	0.9346
Mn:U	27-group	0.7395	0.5165
128:1	238-group	1.1713	0.5924
	MCNP	0.9194	0.5996
Cr:U	27-group	0.7941	0.9006
235:1	238-group	0.9451	0.9675
	MCNP	0.8857	0.9639
Na:U	27-group	0.8013	0.8619
1450:1	238-group	0.9620	0.8997
	MCNP	0.9570	0.9051

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}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}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}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.

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